# SECOND DRAFT TECHNICAL REPORT (REVISION 1)

# COMPILATION OF EXISTING STUDIES ON SOURCE APPORTIONMENT FOR PM<sub>2.5</sub>

Contract No. 68-D-02-061 Work Assignment 1-05

for

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#### **ACKNOWLEDGEMENTS**

The authors wish to sincerely thank the following people for providing articles, input, and comments to this document.

William Battye, EC/R Incorporated

Jeffrey R. Brook, Environment Canada

Matthew P. Fraser, Rice University

Philip K. Hopke, Clarkson University

Donna M. Kenski, Lake Michigan Air Directors Consortium

Eugene Kim, Clarkson University

Timothy V. Larson, University of Washington

Charles W. Lewis, U.S. Environmental Protection Agency

Joellen Lewtas, U.S. Environmental Protection Agency

Russell W. Long, U.S. Environmental Protection Agency

John M. Ondov, University of Maryland College Park

Seungshik Park, University of Maryland College Park

Richard L. Poirot, Vermont Department of Environmental Conservation

Mei Zheng, Georgia Institute of Technology

#### **ABBREVIATIONS**

AAAR American Association for Aerosol Research
AIRS Aerometric Information Retrieval System
ATAD Atmospheric Transport and Diffusion model

CAPITA Center for Air Pollution Impact and Trend Analysis (Washington Univ.)

CAPITA Monte Carlo Back trajectory model developed by CAPITA

CASTNET Clean Air Status and Trends Network
CMB Chemical Mass Balance model
DFPSS Dual fine particle sequential sampler
DMPS Differential mobility particle sizer

EC Elemental carbon

EMPACT Environmental Monitoring for Public Access and Community Tracking

EPA United States Environmental Protection Agency

GC/MS Gas chromatography/mass spectroscopy

GIS Global information systems

HQ Headquarters

HRM Houston [Texas] Regional Monitoring

HTM Hemispheric Trajectory Model

HYSPLIT Hybrid Single-Particle Lagrangian Integrated Trajectory model

ICP Inductively coupled plasma

IMPROVE Interagency Monitoring of Protected Visual Environments

LADCO Lake Michigan Air Directors Consortium

MDL Method detection limit
ME Multilinear Engine
MS Mass spectrometry

NESCAUM Northeast States for Coordinated Air Use Management

NVOM Non-volatile organic matter

OC Organic carbon

PAH Polycyclic aromatic hydrocarbon PCA Principal Component Analysis

PM Particulate matter

PM<sub>2.5</sub> Particulate matter less than 2.5 micrometers in diameter

PMF Positive Matrix Factorization model

PMF3 Positive Matrix Factorization, 3-dimensional PSCF Potential Source Contribution Function model

RPO Regional Planning Organization RTA Residence-Time Analysis model

SEARCH Southeastern Aerosol Research and Characterization

SEM Scanning electron microscope/microscopy

SIP State implementation plan
STAR Science to Achieve Results
TexAQS Texas Air Quality Study
TMB Trajectory Mass Balance mo

TMB Trajectory Mass Balance model UNMIX Source apportionment model VOC Volatile organic compound

XRF X-ray fluorescence

#### **EXECUTIVE SUMMARY**

In 1997, the U.S. Environmental Protection Agency (EPA) revised its particulate matter standards to include an annual standard for fine particulate matter of  $15 \mu g/m^3$  and a 24-hour standard of  $65 \mu g/m^3$ . At this time, EPA also issued rules related to an expanded monitoring network for PM<sub>2.5</sub> so that cities could evaluate their compliance with the new standards. In addition to urban areas, large national parks and wilderness areas (Class I areas) also have visibility requirements, which are directly linked to particulate matter levels.

In any particular urban or Class I area, understanding the sources of ambient particulate matter has become increasingly important. While local sources can be monitored and subjected to local control regulations, particulate matter that has been transported into the region cannot be easily monitored or controlled. Research in the late 1990s began to indicate that transport could be a major source of  $PM_{2.5}$  in some areas. Identifying and quantifying the sources of particulate matter affecting a particular location through source apportionment methods is now an important component of the information available to decision makers when evaluating the new standards.

EPA is now considering a rule based on  $PM_{2.5}$  transport in the eastern part of the United States. As part of a research and preparation program, this literature compilation is designed to summarize where some of the source apportionment research has been conducted and its general findings. EPA staff selected the literature in this compilation as representative of the key source apportionment research, focusing primarily (but not exclusively) on recent studies using the PMF and UNMIX source apportionment models in the eastern U.S.

This compilation reviewed 27 articles in detail. Additionally, there is a survey of recent and current work from conference and workshop presentations. The results of these studies are presented here in unified tabular forms. Specifically, the tables summarize the receptor locations, study methods and study findings, and the source categories identified. From these tables, general observations and recommendations are made to guide future research.

The results are generally comparable when comparable data sets are used. However, methodologies vary considerably among the studies. In addition, the primary goals of apportioning total PM or visibility are quite different. Hence, there are difficulties with making direct comparisons.

The commonly identified source categories include: secondary sulfate/coal burning (sometimes over 50 percent of the mass), secondary organic carbon/mobile sources, crustal sources, biomass burning, nitrate, industrial, smelters and metal processing, and sea salt in coastal regions.

Frequently, the smaller sources and the mobile sources appear as combinations of sources. The combined sources are usually those that would naturally affect the receptor in tandem, such as a mobile — road dust or road salt combination. The main tools for apportioning the sources rely on variations in source strength to separate the sources; hence, sources acting in tandem cannot be separated. Consequently, while separating the mobile sources into diesel- and

gasoline-based emissions is clearly a goal for several of the studies, the success depends on either additional data or analyses.

This compilation is intended to guide future research efforts. The tools and methodologies have passed the proof-of-concept stage and are now being used to understand the ambient composition of the PM for sites across the eastern United States and the spatial relationship of sources to the receptor. However, this should not be taken as an indication that the tools and methods have been finalized. Important method development is needed and continues. There is need for improvement in the main source apportionment tools, in particular, the error estimates and methods for identifying the number of sources. Very little research has been done to date on identifying the sources from the tool output. The methods for identifying the source regions are evolving along two major paths initiated by Richard L. Poirot and Philip K. Hopke. Finally, no methods for the quantification of transport versus local within a given source category have yet been published.

#### 1.0 INTRODUCTION

# 1.1 Background and Purpose of Study

In 1997, the U.S. Environmental Protection Agency (EPA) revised its particulate matter standards to include an annual standard for fine particulate matter (defined as less than 2.5 micrometers in diameter and abbreviated as  $PM_{2.5}$ ) of 15  $\mu g/m^3$  and a 24-hour standard of 65  $\mu g/m^3$  (EPA, 1997a). At this time, EPA also issued rules related to an expanded monitoring network for  $PM_{2.5}$  so that cities could evaluate their compliance with the new standards (EPA, 1997b). Challenges to the regulations, resolved by the Supreme Court in 1999, delayed implementation, but many cities now face compliance requirements within the next few years. In addition to urban areas, large national parks and wilderness areas (called Class I areas) also have visibility requirements, which are directly linked to particulate matter levels.

In any particular urban or Class I area, determining the sources of ambient particulate matter has become increasingly important as the regulations tighten and become more focused on PM<sub>2.5</sub>. While local sources can be monitored and are subject to local control regulations, particulate matter that has been transported into the region cannot easily be monitored or controlled. Research in the late 1990s began to indicate that transport could be a major source of PM<sub>2.5</sub> in some areas. Quantifying the sources of particulate matter in a particular region (source apportionment) is now important for decision makers when evaluating compliance with the new standards.

EPA is now considering a rule based on PM<sub>2.5</sub> transport in the eastern part of the United States. As part of a larger program of research and preparation, this literature compilation is designed to summarize where some of the source apportionment research has been conducted and its general findings. EPA staff selected the literature in this compilation as representative of the key source apportionment research, focusing primarily (but not exclusively) on recent studies using the PMF and UNMIX source apportionment models in the eastern U.S.

This compilation report is divided into two sections: written narrative and summary tables. The written narrative consists of a discussion of general findings and recommendations derived from review of the articles and presentations. The summary tables, located in Appendix A, include information from the reviewed articles and presentations as the authors of the articles presented the information.

# 1.2 Summary of Key Source Apportionment Tools

The main goal for the studies considered is to describe and quantify the major source categories contributing to the observed concentrations of fine particulate matter in the atmosphere. This is done by modeling the  $PM_{2.5}$  mass concentration and 10 to 30 constituent species as a mixture from the major sources that varies from day-to-day. At least initially, it is assumed that the sources contributing to the  $PM_{2.5}$  contribute to the observed species concentrations with approximately fixed ratios. The list of the ratios of the species mass to the total mass contributed by the source is referred to as a source profile. It is also assumed that the

sources can be identified through these ratios, the associated time series (the day-by-day apportioning of the constituent species mass), and the magnitude of the source.

Below is a brief review of the main tools and methods used in the analyses. The tools are grouped into primary tools that are used to apportion the mass and secondary tools that are used to analyze the primary tool output.

# **Primary Source Apportionment Tools**

Positive Matrix Factorization (PMF or PMF2) is a model developed by Penti Paatero. This source apportionment model uses constrained, weighted least squares estimation to apportion the species masses. The input data include the species masses and the uncertainties associated with each measurement. The main outputs are the source profiles and the associated time series (the day-by-day apportioning of species mass). Secondary output includes various model diagnostics.

Multilinear Engine (ME) and Positive Matrix Factorization (3-dimensional) (PMF3) were also developed by Penti Paatero. These source apportionment tools generalize the standard PMF model. The ME model also allows for known constraints and an even broader range of models. The output for both is similar to the PMF output.

UNMIX was developed by Ronald Henry. This model apportions the data based on the "edges" produced in the data when one or more of the sources do not significantly contribute to the total mass of any species being modeled. Formally, the model uses an additional assumption that for each source there are periods when the source does not significantly contribute to any of the modeled species. However, this criterion is required to ensure that the solution is unique for all models that do not use known emissions profiles. In addition to the source profiles and the associated time series, this software also includes plotting tools for identifying outliers and diagnostics for deciding how many sources should be modeled. The latter depend on the species to be modeled.

Chemical Mass Balance (CMB) was developed by John Watson and associates. This model apportions the mass using historical emission source profiles that are assumed known and weighted regression methods. The output does not include the source profiles, since they are required inputs. [See Chow and Watson (2002) for a survey of applications of this model.]

# **Secondary Tools and Methods**

There are a variety of secondary tools and methods used in conjunction with the source apportionment tools to investigate and possibly refine the source apportionment. Chief among these are methods that pair the source apportionment output with meteorological data.

The simplest pairing is a pollution rose, which is a plot that indicates the source strength by wind direction. There are actually several varieties of these plots, but they all convey the same general information. These plots are useful in locating the general direction of a local source. They may also be useful in identifying sources that are dependent on wind speed. The meteorological data for these plots usually originate from the nearest weather station. Generally,

this means that the receptor and the meteorological data are not from co-located instruments, which can severely limit the usefulness or accuracy of these plots.

The second pairing of the source apportionment data with meteorological data is based on back trajectory methods. In this case, the source apportionment output is paired with the output from a meteorological model that indicates a likely path back in time for a packet of air arriving at the receptor location during the sampling period. Inferences on the source location(s) are made by comparing the paths that correspond to high source strengths with all paths generated from the modeled period and/or the paths that correspond to low source strength periods. Since the inference is generally made through a probabilistic framework, the output is sometimes referred to as a probability field. These methods are also referred to as (conditional) ensemble back trajectory methods.

The two most common meteorological models used in the studies surveyed for generating the back trajectories are the Atmospheric Transport and Diffusion (ATAD) model and the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. ATAD can use raw meteorological data or gridded output from a meteorological preprocessor. HYSPLIT requires gridded preprocessed data. The choice of the preprocessed data is study-specific.

Subtle differences exist among the explicit methods, the meteorological models, and the meteorological data used among the studies, even among studies by the same author. However, in the reports surveyed, the results generally are used qualitatively. At this level, the various methods and models yield comparable interpretations. This issue may need to be reassessed when quantitative methods are developed for estimating the transport from the indicated source regions.

# 2.0 DISCUSSION OF STUDIES

This compilation reviewed 27 articles in detail, surveyed presentations of recent work, and scanned a range of other articles. One of the key areas of interest was the locations of receptors that had been studied. Table A-1 is a detailed summary showing where the receptors for each study were located, including latitude and longitude information when available. Figure 1 is a map showing the coverage of these studies, and Table 1 gives the location names. This survey focused on the eastern and midwestern portion of the United States. Figure 2 shows pie charts of the various apportionment results.

Table A-2 is a detailed summary of the methods and findings in each of the published studies. Table A-3 is a similar summary of recent presentations. Below is a general discussion of the common themes and areas of difference between the studies, as well as areas of research that are on-going or proposed at this time.

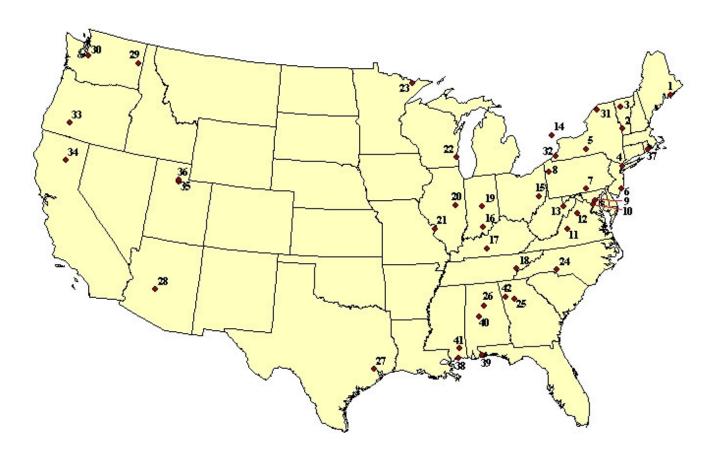


Figure 1. Map of source apportionment studies involving PM<sub>2.5</sub>

Table 1. Location of source apportionment studies involving PM<sub>2.5</sub>

Label	Location or Nearest City	Label	Location or Nearest City	Label	Location or Nearest City
1	Acadia National Park, ME	15	Quaker City, OH	30a-c	Seattle, WA
2	Lye Brook Wilderness, VT	16	Livonia, IN	31	Potsdam, NY
3a-d	Underhill, VT	17	Mammoth Cave National Park, KY	32	Stockton, NY
4	Bronx, NY	18	Great Smoky Mountains National Park, TN	33	Crater Lake National Park, OR
5	Connecticut Hill, NY	19	Indianapolis, IN	34	Lassen Volcanic National Park, CA
6a-b	Brigantine National Wildlife Refuge, NJ	20	Bondville, IL	35	Salt Lake City, UT
6c	Brigantine, NJ	21	St. Louis, MO	36	Bountiful, UT
7	Arendtsville, PA	22	Milwaukee, WI	37	Narragansett, RI
8	M.K. Goddard, PA	23	Boundary Waters Canoe Area, MN	38	Gulfport, MS
9	Fort Meade, MD	24	Charlotte, NC	39	NW of Pensacola, FL
10a-d	Washington, DC	25a-c	Atlanta, GA	39	Pensacola, FL
11	Jefferson/James River Face Wilderness, VA	26	Birmingham, AL	40	Centreville, AL
12	Shenandoah National Park, VA	27	Houston, TX	41	Oak Grove, MS
13	Dolly Sods/Otter Creek Wilderness, WV	28	Phoenix, AZ	42	Yorkville, GA
14	Toronto, ON	29	Spokane, WA		

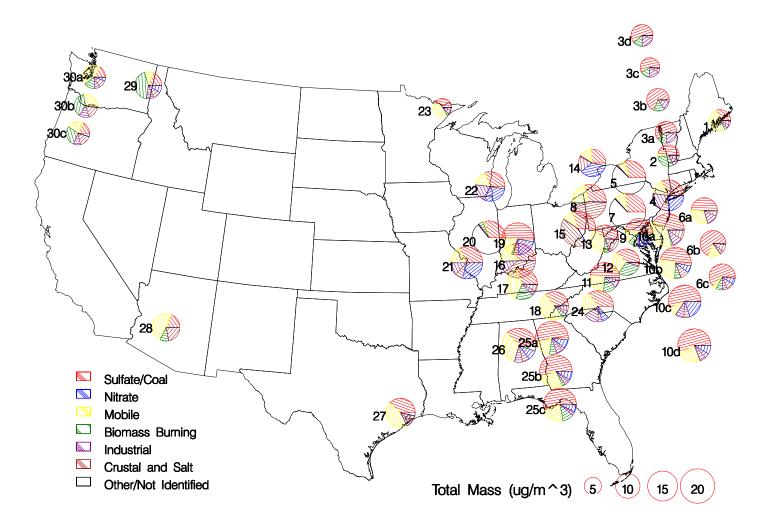


Figure 2. Pie charts of the source apportionment results. (Some charts moved for clarity. See Tables 1 and A-4 for the site names and study references.)

#### 2.1 Overview of the Results

The general results from many of the studies were found to be similar. A few receptors were studied repeatedly, such as Underhill, Vermont, and Brigantine, New Jersey. As the methodologies have become more mature, there was a trend toward research on a broader set of locations and the evaluation of regional trends. This trend toward broader site selection is also driven by regulations that encompass broad regional areas, such as the regional haze rule and the anticipated transport rule. The trend is also driven by the fact that source apportionment is still quite specialized and is generally only supported by Regional Planning Organizations and larger organizations that have regional or national level concerns.

Table A-4 is a matrix that presents an overview of the sources found in the studies in order of location. The sources identified are grouped into seven categories: sulfate/coal, mobile, nitrate, biomass burning, industrial, crustal and salt, and other/not identified. Each of these source categories is discussed below.

Note that in Figure 2 the results from neighboring sites are generally quite similar. The only exceptions are from the CASTNET sites: Connecticut Hill, New York; M.K. Goddard, Pennsylvania; Quaker City, Ohio; and Bondville, Illinois. Also note that none of the study apportioned masses have been adjusted to account for the fact that different seasons may not be equally represented. Sources that are strongly seasonal, in particular both the sulfate and nitrate sources, may be unequally represented in the apportionment means. For example, if a study covered a 16-month period from January of one year to April of the following year, the total mass apportioned to nitrate may in fact represent a two-year total (since nitrate is generally observed in the winter), while the mass apportioned to the sulfate may represent a one-year total (since sulfate is usually higher in the summer).

#### Sulfate/coal

Secondary sulfate/coal was identified as the largest or one of the largest sources in nearly every study, often consisting of over 50 percent of the source of PM<sub>2.5</sub> at some locations during some seasons. In a few cases, there was a known local source of sulfate, but most of the eastern studies (in conjunction with back trajectory analysis) pointed to coal-fired power plants in the Midwest. The studies with multiple years of data also tended to identify a winter and summer signature to the sulfate source, with the summer version apportioning more mass. The studies speculate that the two profiles represent two extremes in the atmospheric chemistry between the source regions and the receptor. Note that the source category is often referred to by its dominant species, sulfate, but the "sulfate source" is often associated with significant amounts of organic carbon and is usually the single largest source of selenium and other trace elements.

#### Mobile

Secondary organic matter/mobile sources are also a major source for nearly all sites. As with sulfate, the source is sometimes named after the dominant species since it is often formed through a secondary process in the atmosphere rather than being emitted directly. This case is

even further complicated by the fact that the particulate organic carbon is itself a mix of many species that are not usually measured separately.

Only a few studies are able to separate the mobile source into gasoline sources and diesel sources. It may be that additional speciation of the organic carbon or thermal fractionization of the organic carbon is needed for the tools to separate consistently the two major components of the mobile source category. Both approaches have been tried with some success. However, the fractionization method is limited by the lack of source emission profiles with comparable thermal fractionization of the particulate carbon. Hence, the results cannot be verified against known profiles. The chemical speciation of the carbon can be expensive. Of the studies compiled, only one study measured an organic carbon subspecies. The purpose of the particular study, however, was to apportion wood smoke. Hence, the additional species measured was a wood smoke tracer. Further studies using chemical speciation of the organic carbon are planned.

#### **Nitrate**

Among the eastern sites, a nitrate dominated source is also found to be a major source. It is often the second largest source. The back trajectory analyses sometimes show an association with agricultural areas that would have high ammonia emissions.

Because nitrate is semi-volatile and is much more prevalent in the winter, the source apportionment tools should separate out the nitrate as a separate source even when it is a component of other sources. Recall that the tools assume that the species-to-species ratio is fixed for any source. This is generally not true for nitrate, so the models should separate this component of the source as a separate source. Further problems measuring nitrate within the IMPROVE network have resulted in exclusion of this species as a fitting species for many of the sites. Hence, the "nitrate" source may be one of the unidentified sources for sites without a nitrate component.

# **Biomass burning**

The biomass burning category includes the wood smoke and forest fire categories identified at several sites. The size of the source varies considerably from site to site, but usually as expected (e.g., larger in rural areas and in the northwest).

Sometimes, this category also includes the fireworks noted by several studies. This is because the source is characterized by organic carbon and potassium. Usually an explicit reference to fireworks is based on a 4th of July spike in the source strength, but may also be supported by trace metals, particularly copper, found in the profile. In any case, the source profiles are similar enough and the source strength small enough that the models do not generally separate biomass burning from fireworks.

#### Industrial

This category includes a variety of small sources characterized by elemental carbon and trace metals, such as smelters and incinerators that may or may not have been found at the various sites. Since this roughly fits the leading characteristics of oil burning (sulfate, carbon,

Ni, and V), any of the oil burning sources are included in this category in the pie charts in Figure 2.

Frequently, the industrial sources are associated with known local sources or, in the case of the northeast, known smelters in Canada. These sources also tend to be distinctive enough for the models to separate them into several small sources within a site.

# **Crustal and Salt**

The crustal source category is identified for all sites, but is usually small, 0.1 to  $1.5 \,\mu\text{g/m}^3$ . There are three notable rural exceptions: M.K. Goddard, Pennsylvania; Quaker City, Ohio; and Livonia, Indiana, each with 7.8  $\mu\text{g/m}^3$  or more. The Phoenix site is also apportioned a larger crustal source,  $2.8 \,\mu\text{g/m}^3$ .

Various small sources with a salt component are also included in this category for the pie charts. Hence, road dust—road salt combinations are shown in this category rather than the mobile category. Marine salts, which are always quite small, are also included in this category. Industrial-salt sources, however, are in the industrial category.

# Other/Not Identified

Four of the six CASTNET sites (Arrentsville, Pennsylvania; Connecticut Hill, New York; Quaker City, Ohio; and Bondville, Illinois) have large (> 3  $\mu g/m^3$ ) unidentified sources. The particular study was concerned with light extinction; since these were not significantly associated with light extinction, it was not felt necessary to identify those sources. Otherwise, sources greater than 1  $\mu g/m^3$  are identified. The remaining miscellaneous sources are generally under 1  $\mu g/m^3$  also.

While the majority of the literature was focused on sites in the eastern and midwestern United States, several studies that were reviewed were based in western locations, including Arizona (Lewis, et al., 2002), Utah (Long, 2002), and Washington (Maykut, et al., 2003). The general types of sources in the West were similar to those of eastern locations with some general differences in the source proportions. Mobile sources tended to be a larger portion and secondary sulfate a smaller portion of the total mass compared to eastern locations. Vegetative burning also tended to have a greater influence in western sites, likely due to forest fires.

# 2.2 Source Locations and Time Series Analyses

This study concentrated on the source apportionment models of Positive Matrix Factorization (PMF), its variations, and UNMIX. PMF and UNMIX were used as the sole source apportionment tool and/or as a check on the results of the other model. More importantly, nearly all studies agreed that source apportionment models cannot stand alone for many of the desired uses. In fact, additional supporting evidence is frequently needed to complete the source identification process. Thus, the models are usually used in conjunction with other tools, commonly back trajectory analyses via a meteorological model such as the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.

All back trajectory analyses for the eastern sites associate the sulfate with the Ohio River Valley area. Industrial sources are also frequently associated with known source areas. Several studies noted transport across the Canadian border, specifically sulfates from the midwestern United States into Canada, and smelter emissions from Canada into the northeastern United States. There are plans to use the back trajectory data to quantify the transport; however, none of those studies are complete yet.

All of the studies looked at long-term averages and most looked at seasonal (3-month) averages. There was very little analysis of daily or weekly events, with a few exceptions. (For the most part, the studies considered are motivated by long-term concerns, such as trends in regional haze.) Lee, et al., (2003a) followed up on a crustal source by identifying several days that were possibly influenced by Saharan dust. Coutant, et al., (2002) mention the influence of fireworks on July 4-5 in Houston, Texas. Long (2002) studied a particular event (2002 Winter Olympics) and documented changes in the source proportions (mobile sources were higher) and temporal changes (mobile sources were evenly distributed instead of exhibiting a diurnal pattern).

In several cases where datasets covering very long time periods were evaluated, reductions in emissions could be seen for power plants (Poirot, et al., 2001), fuel oil (Lee, et al., 2003a), and smelters (Battye, 2002). These were attributed to increased emission controls, fuel switching (e.g., from oil to natural gas), and meteorological conditions (e.g., warmer winters in the late 1990s).

# 2.3 Methodologies and Technical Approaches

The technical approach varies significantly among the various studies. Some studies, through preplanned additional data collection, have also used tools such as scanning electron microscope analysis of the particulate matter or specialized tracers to gain a greater understanding of specific PM<sub>2.5</sub> sources. Typically, however, the data used are very similar to data from IMPROVE or IMPROVE protocol sites or more recently from EPA's Speciation Trends Network, with a few super sites having specialized data.

PMF, its variations, and UNMIX represent very different approaches to source apportionment. Data preprocessing for missing data and identification of outliers is not standardized. Profile interpretation is essentially a matter of "expert opinion." Even the derivation and processing of the back trajectories varies significantly among the studies surveyed.

Where both models have been used, PMF has been used to model more sources than UNMIX. However, PMF is typically used to model more species, so it should be able to identify more sources. This is probably driven by the fact that multiple modeling steps are sometimes required to model a large number of species with UNMIX. The results then need to be merged into a single solution. PMF is generally not used in this manner, except for apportioning the total mass.

The preprocessing of the data for use in the models is dependent on the amount of data available and the particular study goals. For example, if long-term trends are a part of the study

goals, then isolated events are sometimes screened. Missing data, or rather incomplete data, are sometimes handled by data imputation and sometimes deletion of the data. Below minimum detection data are fairly consistently handled by MDL/2 substitution.

The interpretation of the numerical results requires human judgment; hence, study results are dependent on the modeler's skill. Many studies use multiple rounds of review to avoid or lessen the impact of this fact. Coutant, et al., (2002) uses profile matching algorithms to match the output with Speciate profiles, in order to create a more objective profile identification process; however, the algorithms and Speciate database can only be used as an additional round of review at this time, not as a final product.

Analyses of the time series output, particularly back trajectory methods, are frequently being used to aid interpretation. However, this adds an additional layer of divergent methods and models. ATAD and HYSPLIT are the two most common models used to generate the individual back trajectories. The methods for implementing these models vary in the choices of starting times and heights and in other technical aspects. The processing of the back trajectories also varies considerably in the definition of high and low day source strength, the base unit used from the trajectories (hour or number of end points), the metric used to measure the relative likeliness of the source location, and the contouring methods. Some consistency has been reported (Gepart; Kenski) among the various final results from divergent intermediate steps, but preferred methodologies have not been identified.

Consequently, one should not expect identical results from different studies even when the same base data are used. Consistent results for the major source categories are seen as proof-of-concept for all involved, and have been used as an internal check on procedures within studies. However, at this time, there are no clearly preferred models or sets of methodologies.

#### 2.4 Current Issues

As more researchers continue to apply the tools, new ideas are emerging on how the tools can best be used. Several ideas are highlighted below, mainly because they directly attack issues of interest.

# Determining the gas/diesel split in the mobile component

This issue has already been mentioned, along with two of the methods for attempting to measure the split.

# Determining the number of sources to be modeled

The method used to determine the number of sources to be modeled varies significantly from study to study, even within researcher. Guidance for this issue, together with various data handling issues, will be needed before considering routine use of the models for regulatory purposes.

# **Determining the error associated with the model**

Both PMF and UNMIX produce error estimates. UNMIX uses a boot-strapping method for making error estimates. The error estimates are usually comparable to PMF error estimates. PMF gives estimates that are based on the assumption that either the profiles or the source strength estimates are known and error free, and that the uncertainty measurements are correctly specified and the number of sources is correctly specified. The first of these assumptions is, of course, false; in fact, it runs contrary to the purpose of the model. In Coutant, et al. (2002), the difference between the sulfate and elemental sulfur apportionments were compared. For the IMPROVE sites studied, the sulfate measurement is consistently (within measurement error) three times the sulfur measurement as expected under the hypothesis that all the sulfur is present in the form of sulfate. Hence, the apportionments should always have a ratio of three. This was found to be generally true, but the errors were much larger than indicated by the PMF or UNMIX output. Hence, this study switched to using an estimate of the errors based on the difference between the sulfate and sulfur apportionments rather than using the PMF or UNMIX output.

# 2.5 Proposed and On-going Studies

Most of the published literature did not discuss proposed or on-going studies. However, the presentations and direct communication with the authors did reveal some on-going work.

Poirot and Wishinski (2001) plan to conduct further research in determining specific events, such as influx of Saharan dust into Brigantine. They have also been conducting analyses to better differentiate between sea salt and road salt. Separately, Poirot mentioned improving comparison of PMF and UNMIX and attempting to refine the mobile (diesel versus gasoline) and sulfate (acidic versus neutral) sources (Poirot, 2003).

Several researchers are beginning to conduct comparison studies between different models, including comparison of CMB and PMF in the Houston area (Fraser). Similar on-going and proposed studies were presented at the AAAR meeting in October 2002. Comparisons of PMF and UNMIX are being conducted by Eugene Kim, who found that results are similar but not the same, with diesel emissions the most unalike. Philip Hopke is studying Atlanta with the Multilinear Engine as a multilinear receptor model, in conjunction with gaseous species and meteorological data. Seungshik Park is using Supersite PM<sub>10</sub> data in Pittsburgh and Baltimore to compare principal component analysis (PCA) and PMF with HYSPLIT, and his preliminary findings show that PCA and PMF yield similar, but not the same, results (e.g., PCA identified a single steel mill source, while PMF split the source into steel mill and coal combustion from the coking process). Additionally, LADCO plans to have a source apportionment study that includes speciated carbon measurements.

Overall, the research trend is to continue comparison studies of the various models, expand the number of receptors studied, and to use various existing datasets from air quality networks. There is also an on-going trend to investigate the use of other supporting technologies, e.g., comparing the back trajectory models to each other (Kenski). There has been little proposed at this stage to standardize the methodologies of either the source apportionment models or the supporting tools.

#### 3.0 RECOMMENDATIONS

The literature revealed a field of study that is still developing. Large speciated PM<sub>2.5</sub> datasets are just becoming available for most regions of the country. Models have been developed and are now being evaluated against each other and in more regions of the country with diverse sources. The limitations of current source apportionment tools are also just now being understood and researchers are experimenting with the joint use of source apportionment models and other tools.

The recommended approach builds on existing work and also focuses on the eastern United States where the potential rule is expected to apply.

### 3.1 Building from Existing Studies

Following is a list of recommended areas of further research and development that builds from the existing studies, particularly where conflicts need to be resolved and where similarities can be confirmed and leveraged.

- 1. <u>Comparison and convergence of models</u>. If source apportionment is to be used for rule development, at some point there needs to be greater consensus and convergence of the results from the models. Thus, considerable further study needs to be conducted comparing the results of the models and the role of decision-making by the modeler in the results. The findings of these comparisons then need to be incorporated into the models.
- 2. <u>Standard method development</u>. Once the model results are better understood, a standard methodology needs to be developed and tested so that results are more comparable. The challenge will be to avoid inhibiting further research by setting a standard too early; that is, if a standard method is established, there is less incentive for continued experimentation. On the other hand, without a more standard method, results between cities, states, and regions are not easily comparable.
- 3. <u>Integration of analytical techniques</u>. The research clearly indicates that using multiple analytical techniques is the best approach, particularly source apportionment models combined with back trajectory models. There is also potential for other combined techniques, such as satellite data, which are being investigated. Research needs to be conducted in how to better integrate these tools and data sets.
- 4. <u>Stakeholder dialogue</u>. Since the data will be used beyond scientific research, dialogue with the ultimate users of this information (e.g., state and local officials) should be conducted to determine what kind of information they would find useful and in what format. This is likely happening informally at the U.S. EPA level, but should also be conducted at the RPO, state, and city level.
- 5. <u>Timeliness</u>. Currently, all the research is being done on historical datasets primarily from the late 1990s and, to some extent, to 2001. Long (2002) proved the concept that hourly data can be used in UNMIX. Research needs to be conducted into how the models, trajectory

analysis, real-time  $PM_{2.5}$  monitoring, and data presentation can be combined to create source apportionment in near real-time.

6. <u>Study of events</u>. Most research has been on long-term averages. However, time-specific air pollution events may have significant impact on the daily PM<sub>2.5</sub> levels in a particular area, and may be from distant sources. Further source apportionment research into air pollution events is recommended in order to provide more information on specific types of sources, such as major and local forest/agricultural fires (including transport), long-distance crustal transport (e.g., Saharan and Asian dust), stagnated urban air pollution that is transported due to frontal systems, fireworks, and temporary change in traffic and industrial patterns (e.g., Olympics).

#### 3.2 Characterization for the Eastern United States

The above recommendations would all help with source apportionment in the eastern United States. Following are some specific research areas that would help improve characterization of PM sources in the eastern U.S.

- 1. <u>Recognize source apportionment and transport analysis as a primary data use.</u> Source apportionment and transport need to be considered in the network development. Characterizations of the main air flow patterns across the U.S. should be used in choosing site locations.
- 2. <u>Additional analytes</u>. Even limited speciated carbon measurements could improve the source resolution of the source apportionment techniques. Co-located meteorological data could be very useful also. Evaluate the current choice of chemical species, such as VOCs, to determine if any others should be monitored and used as inputs to improve source identification, for example, diesel- versus gasoline-based mobile sources.
- 3. <u>Geographic based "source atlas.</u>" Building from Toxics Release Inventory and state emission inventory data, one recommendation is to create an electronic source atlas using GIS technologies that could be used to better understand where potentially large sources or source classes are located. Additional datasets would also need to be created to include event non-point sources such as biomass burning and dust.

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#### 4.2 Website Presentations

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# 4.3 Meeting Presentations

Select presentations at the American Association for Aerosol Research (AAAR) meeting in Charlotte, North Carolina, October 2002.

- 1. Park, S., Pancras, P., Chang, Y., Catino, D., Gazula, S., and Ondov, J. "Investigation of Sources with Highly Time-Resolved Aerosol at the Baltimore Supersite Using PMF." Seung Park, Patrick Pancras. (Poster PA 2-01.)
- 2. Park, S., Pancras, P., Gazula, S., and Ondov, J. "Sources of Elemental Aerosol Constituents in Pittsburgh using PMF of Highly Time-Resolved Data." (Poster PA 2-05.)

Presentations at the LADCO Trajectory/Source Apportionment meeting, in Des Plaines, Illinois, October 22, 2002. Available at http://www.ladco.org/monitor/trajectory\_oct02.

- 1. Fraser, M. "Comparison of CMB and PMF for Source Attribution of PM2.5 in Houston."
- 2. Gebhart, K. "Trajectory/Wind Field Comparisons and Initial Trajectory Mass Balance Results."
- 3. Kenski, D. "Comparing LADCO trajectory ensembles with Vt.DEC trajectory ensembles for Lye Brook (both using ATAD trajectories) *and* Comparing ATAD and HYSPLIT trajectories for high source contribution days at Lye Brook."

4. Kenski, D. "CMB Analysis of Urban PM2.5 Data."

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# **APPENDIX A:**

**SUMMARY TABLES** 

Table A-1. Full description of location of source apportionment studies involving PM<sub>2.5</sub>

Reference	Site Name or Region	Location or Nearest City	Latitude	Longitude	Elevation
Battye 2002	Northeast Border	Kejimkujik, NS St. Anicet, PQ	See Note 1		
		Acadia National Park, ME			
		Cranmore Mountain, NH			
		Lye Brook Wilderness Area, VT			
		Lake Champlain Basin, VT/NY			
		Underhill, VT			
	Mid-Atlantic/	Ringwood, NJ			
	Appalachian	Brigantine National Wildlife Refuge, NJ			
		Philadelphia, PA			
		Baltimore, MD			
		Washington, DC			
		Shenandoah NP, VA Jefferson National Forest, VA			
		Dolly Sods, WV			
	Great Lakes	Egbert, ON			
	Great Lakes	Simcoe, ON			
		Toronto, ON			
		Connecticut Hill, NY			
		Goddard, PA			
		Quaker City, OH			
		Cleveland, OH			
		Livonia, IN			
		Indianapolis, IN			
		Detroit, MI			
		Bondville, IL			
		Chicago, IL			
		Milwaukee, WI			
		Boundary Waters, MN			
	Western Border	Mount Rainier, WA			
		Glacier National Park, MT			
Chen et al. 2002	Fort Meade	Baltimore-Washington corridor	39.10°N	76.74°W	46 m

Reference	Site Name or Region	Location or Nearest City	Latitude	Longitude	Elevation
Chow & Watson 2002		San Jose and San Carlos, CA San Joaquin Valley, CA Fresno, Bakersfield, Kern Nat'l Wildlife Refuge, CA Santa Barbara, CA Los Angeles area, CA Imperial Valley, CA Las Vegas Valley, NV Front range region, CO Chicago, IL Grand Canyon National Park Mt. Zirkel Wilderness Area, CO Lower Fraser Valley, BC Toronto and Egbert, ON (+ 7 other international sites)	See Note 1		
Coutant et al. 2002 See Note 3	IMPROVE Sites  CASTNET Sites	Acadia National Park, ME Boundary Waters Canoe Area, MN Brigantine National Wildlife Refuge, NJ Dolly Sods Wilderness Area, WV Great Smoky Mountains National Park, TN Jefferson/James River Face Wilderness Area, VA Lye Brook Wilderness Area, VT Mammoth Cave National Park, KY Shenandoah National Park, VA Washington, DC Arendtsville, PA Bondville, IL Connecticut Hill, NY Livonia, IN M.K.Goddard, PA Quaker City, OH	44.38 °N 47.95 °N 39.47 °N 39.11 °N 35.63 °N 37.67 °N 43.15 °N 37.13 °N 38.52 °N 38.52 °N 38.92 °N 40.05 °N 42.40 °N 38.53 °N 41.43 °N 39.94 °N	68.26 °W 91.50 °W 74.45 °W 79.43 °W 83.94 °W 73.13 °W 86.15 °W 77.03 °W 77.31 °W 88.37 °W 76.65 °W 86.26 °W 80.14 °W 81.34 °W	150 m 524 m 5 m 1158 m 815 m 280 m 1006 m 248 m 1098 m 16 m 268 m 211 m 505 m 298 m 387 m 376 m

Reference	Site Name or Region	Location or Nearest City	Latitude	Longitude	Elevation
Coutant et al. 2003	Bronx Garden	Bronx, NY	40.86 °N	73.88°W	NA
See Note 3	Blair Street	St. Louis, MO	38.66 °N	90.20 °W	NA
	Aldine Road	Houston, TX	29.90 °N	95.33 °W	NA
	McMillan Site	Washington, DC	38.88 °N	77.03 °W	16 m
	SE Region HQ	Milwaukee, WI	43.06 °N	87.91 °W	NA
	_	Birmingham, AL	33.55 °N	86.82°W	179 m
	Garinger High School	Charlotte, NC	35.24 °N	80.79 °W	230 m
		Indianapolis, IN	39.81 °N	86.11 °W	244 m
Kim and Hopke 2003 See Note 4		Washington, DC	38.55 °N	77.00°W	30 m
Kim et al. 2003a See Note 4	Jefferson Street	Atlanta, GA	33.78 °N	84.41 °W	275 m
Kim et al. 2003b See Note 4	Jefferson Street	Atlanta, GA	33.78 °N	84.41 °W	275 m
Kim et al. 2002a See Note 4	Jefferson Street	Atlanta, GA	33.78 °N	84.41 °W	275 m
Kim et al. 2002b See Note 4	Beacon Hill	Seattle, WA	47.34°N	122.18°W	99 m
Kim et al. 2002c See Note 4	Rockwood	Spokane, WA	47.67°N	117.41 °W	NA
Lee, JH et al. 2003a		Brigantine National Wildlife Refuge, NJ	39.47°N	74.45 °W	9 m
Lee, PKH et al. 2003b	Gage Building	Toronto, ON	43.66°N	79.39°W	17 m
Lewis et al. 2002 See Note 4	3847 W. Earl Dr.	Phoenix, AZ	33.81 °N	112.24°W	307 m
Liu et al. 2003a See Note 4		Potsdam, NY Stockton, NY	44.67°N 42.30°N	74.99 °W 79.36 °W	NA NA
Liu et al. 2003b		Crater Lake National Park, OR	42.90 °N	122.14°W	1981 m
		Lassen Volcanic National Park, CA	42.90 N 40.54 N	122.14 W 121.58 W	1798 m
Long 2002	EMPACT Hawthorne	Salt Lake City, UT	40.76°N	111.89°W	1300 m
See Note 3	STAR Bountiful	Bountiful, UT	40.89°N	111.88°W	1344 m
Lowenthal & Rahn 1989 See Note 4		Narragansett, RI	41.45°N	71.45 °W	NA
Maykut et al. 2003 See Note 4	Beacon Hill	Seattle, WA	47.34°N	122.18°W	NA

Reference	Site Name or Region	Location or Nearest City	Latitude	Longitude	Elevation
Poirot et al. 2001		Underhill, VT	44.53 °N	72.86 °W	400 m
Polissar et al. 2001		Underhill, VT	44.53 °N	72.86 °W	400 m
Rahn & Lowenthal 1984 See Note 4		Narragansett, RI Underhill, VT Barrow, AK (+3 in Europe)	41.45 °N 44.53 °N 71.30 °N	71.45 °W 72.86 °W 156.78 °W	NA 400 m NA
Ramadan et al. 2003	3847 W. Earl Dr.	Phoenix, AZ	33.81 °N	112.24°W	307 m
Ramadan et al. 2000	3847 W. Earl Dr.	Phoenix, AZ	33.81 °N	112.24°W	307 m
Song et al. 2001		Washington, DC Brigantine, NJ Underhill, VT	38.55 °N 39.42 °N 44.53 °N	77.00 °W 74.42 °W 72.86 °W	30 m 50 m 400 m
Wishinski & Poirot 1986 See Note 4	Proctor Maple Research Center	Underhill, VT	44.53 °N	72.86 °W	400 m
Zheng et al. 2002 See Note 3	Jefferson Street OLF#8	Atlanta, GA N. Birmingham, AL Gulfport, MS Pensacola, FL NW of Pensacola, FL Centreville, AL Oak Grove, MS Yorkville, GA	33.78 °N 33.55 °N 30.39 °N 30.44 °N 30.55 °N 32.90 °N 30.99 °N 33.93 °N	84.41 W 86.82 W 89.05 W 87.26 W 87.38 W 87.25 W 88.93 W 85.05 W	275 m 200 m 5 m 27 m 45 m 135 m 100 m 395 m
Website Presentations					
Poirot & Wishinski 2001		Brigantine National Wildlife Refuge, NJ	See Note 2		

Reference	Site Name or Region	Location or Nearest City	Latitude	Longitude	Elevation
Meeting Presentations					
AAAR Presentations 2002 Park (1) Park (2)		Baltimore, MD Pittsburgh, PA	See Note 2		
LADCO Presentations 2002 Fraser Kenski (1) Kenski (2)		Houston, TX Lye Brook, VT Chicago, IL Indianapolis, IN Detroit, MI Cleveland, OH Milwaukee, WI St. Louis, MO	See Note 2		

Note 1: Compendium of studies, location names, but not specific latitude and longitude, provided. Note 2: Latitude and longitude not reported in study.

Note 3: Latitude and longitude not reported in study but provided by author directly.

Note 4: Latitude and longitude are estimates from other studies, network locations, or general city location.

Table A-2. Summary of source apportionment studies involving PM<sub>2.5</sub>: Papers and reports

**Study Reference:** Battye, W. (2002). Compendium of existing back-trajectory analyses relating to US-Canada trans-boundary impacts of fine particulate matter and regional haze. Draft Report for U.S. Environmental Protection Agency, Contract No. 68-D-98-006, Work Assignment No. 5-07, September.

No. 5-07, September. <b>Type of Study:</b> Literature review/o	contract research report	Funding Source: U.S. EPA
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): 31 sites within 500 km of U.SCanadian border (see Table 1 for complete list)	Source Apportionment Tool (s): Trajectory analysis only.  Number of Sources/Species: Trajectory analysis only.	Results:  The report presented graphics of back trajectory and probability fields for four regions: northeast border, mid-Atlantic/Appalachian, Great Lakes, and western border
Timeframe: Varied by study; ranged from 1983-2000, but most in mid to late 1990s. Studies used	Sources: Trajectory analysis only.  Methodology: The report collected and reviewed PM <sub>2.5</sub> and regional haze back trajectory results from research studies conducted on sites along the U.SCanadian order. Three types of methods were included: (1) correlation of pollutants to wind speed and direction; (2) four trajectory models for the 20% of the days with the highest pollution concentration and the 20% with the lowest concentration; and (3) three types of ensemble trajectory analysis. Results of these different	The northeast border region generally has low visibility days associated with transport from the west, southwest, or south. High visibility days were generally associated with north and east trajectories, with some low visibility days from the north.
seasonal, annual, and multi-year data.  Frequency: Not specified.  Data Source: Not specified, although citations		In the mid-Atlantic, a number of high concentration/low visibility days in New Jersey appeared to come from the direction of Canada, however, these trajectories also traverse potential source regions in Pennsylvania and New York state. Less information was available for the more southern part of the mid-Atlantic.
for the published studies are provided.  Data Description: Specific to each study, including PM <sub>2.5</sub> , visibility, sulfate, and		In the midwest, low visibility and high sulfate days were generally associated with transport from the south. High visibility days corresponded with winds from the north, though some poor visibility days were associated with winds from the northwest.
specific metals.	methods were compared within 4 geographic regions.  Other Analyses:	In the western U.S., not enough sites were analyzed to identify significant trends, although transport in both directions across the border is likely.
	Trajectory models: Hybrid Single- Particle Lagrangian Integrated Trajectory (HYSPLIT) model, Atmospheric Transport and Dispersion	Temporal Signals: Impacts from Ontario smelters appeared to decline in the late 1980s.
	(ATAD) model, CAPITA Monte Carlo	Transboundary Transport:

**Study Reference:** Battye, W. (2002). Compendium of existing back-trajectory analyses relating to US-Canada trans-boundary impacts of fine particulate matter and regional haze. Draft Report for U.S. Environmental Protection Agency, Contract No. 68-D-98-006, Work Assignment No. 5-07, September.

Type of Study: Literature review/contract research report Funding Source: U.S. EPA

Type of Study. Enclature review	v/contract research report	Funding Source. U.S. El A		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations		
	model, and the Hemispheric Trajectory Model (HTM) model. Ensemble trajectory models: Potential Source Contribution Function (PSCF), Residence-Time Analysis (RTA), and Trajectory Mass Balance (TMB).	In the northeast U.S., several studies identified a Canadian smelter source with emissions that appeared to decline between in 1986 and 1995. Sulfates, wood smoke, and mobile emissions are all transported across the border at some scale. In the midwest, transport of sulfates is indicated across the border from the U.S. to Canada. Further west, transport of sulfates may be occurring from Canada into national parks in the western U.S. as well as transport from the U.S. to Canada.		
		Recommendations: Patterns and constituents of transport seem to have changed over the past 15 years, so it is difficult to compare studies. Joint source apportionment and back trajectory studies should be conducted on more recent data.  Upcoming Studies/Planned Work: None mentioned.		

Study Reference: Chen, L.-W.A., B.G. Doddridge, R.R. Dickerson, J.C. Chow, and R.C. Henry (2002). Origins of fine aerosol mass in Baltimore-Washington corridor: implications from observation, factor analysis, and ensemble air parcel back trajectories. Atmospheric Environment, 36, 4541-4554.

Type of Study: Peer reviewed research

Funding Source: Baltimore Gas and Electric Company and

Type of Study. Teel leviewed les	caren	Potomac Electric Power Company through the Electric Power Research Institute and Maryland Industrial Partnerships; EPA.
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site and Data Specifications  Site Location(s) and Type(s): Fort Meade, MD (suburban)  Timeframe: 1999-2001  Frequency: 24-hour daily averages in July, October, January, and April  Data Source: Special study  Data Description: Total PM <sub>2.5</sub> , speciated PM <sub>2.5</sub> , and trace gases (including NH <sub>3</sub> , HNO <sub>3</sub> , CO, SO <sub>2</sub> , and NO <sub>y</sub> )	Source Apportionment Method  Source Apportionment Tool (s):    UNMIX  Number of Sources/Species:    6 sources and over 30 species.  Sources:    Regional sulfate, Se/sulfate, secondary nitrate/mobile, mobile, Br/K/wood smoke, Cu/Fe/sulfate.  Methodology:    First, an assessment of the trends of the measured species was conducted. Then, the UNMIX model was used with 9 input variables (including mass, sulfate, nitrate, ammonium, organic carbon (OC), elemental carbon (EC), Se, Br, and Cu) to identify 6 sources.  Trajectory analysis was used to develop probability fields for high days and all days (background). These were combined to determine information about the 6 sources.	Results: Fort Meade is representative of the Baltimore-Washington corridor. The annual mean concentration was 13 μg/m³, primarily consisting of sulfate, nitrate, ammonium, and carbonaceous material. Six sources were identified. Regional sulfate (5.28 μg/m³) is from the midwest and moves into Maryland from the north. A second sulfate source (0.93 μg/m³) occurs during stagnant air conditions and represents a local sulfur source. Two signatures are likely the same source (mobile emissions) but with different winter (2.03 μg/m³) and summer (1.77 μg/m³) signatures. Wood
	Other Analyses: HYSPLIT back trajectory analysis	are driven by fewer but stronger events in the winter.  Transboundary Transport:  The correlation between SO <sub>2</sub> and sulfate is weak in any season, which implies transport of SO <sub>2</sub> from distant sources.  Particulate nitrates are higher in the winter.

<b>Study Reference:</b> Chen, LW.A., B.G. Doddridge, R.R. Dickerson, J.C. Chow, and R.C. Henry (2002). Origins of fine aerosol mass in Baltimore-Washington corridor: implications from observation, factor analysis, and ensemble air parcel back trajectories. <i>Atmospheric Environment</i> , <b>36</b> , 4541-4554.				
<b>Type of Study:</b> Peer reviewed re	esearch	Funding Source: Baltimore Gas and Electric Company and		
		Potomac Electric Power Company through the Electric Power		
		Research Institute and Maryland Industrial Partnerships; EPA.		
G'A ID A G 'C' A'				
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations		
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations  Recommendations:		
Site and Data Specifications	Source Apportionment Method			

**Study Reference:** Chow, J.C. and J.G. Watson (2002). Review of  $PM_{2.5}$  and  $PM_{10}$  apportionment for fossil fuel combustion and other sources by the chemical mass balance receptor model. *Energy & Fuels*, **16**, 222-260.

Type of Study: Literature survey

Funding Source: None mentioned

#### **Site and Data Specifications**

# Site Location(s) and Type(s): Over 20 locations of all types in the U.S. (primarily western states) and internationally (see Table 1 for complete list)

#### Timeframe:

1990-1998 with the exception of one study that used data from 1982. All the studies were published between 1995 and 2001.

#### Frequency:

Varied considerably between the 22 studies, including continuous 48 hour sampling, daily 24 hour averages, 12 hour day/night, 3 hour 8 times per day, 6 hour 4 times per day, 6 hour 2 times per day, daily every 6<sup>th</sup> day, and daily every 3<sup>rd</sup> day.

#### Data Source:

Variety of special studies in specific locations.

# Data Description:

PM<sub>2.5</sub> and PM<sub>10</sub> from fossil fuel combustion and other sources. Analytical methods varied between studies.

### **Source Apportionment Method**

Source Apportionment Tool (s):
All studies use the Chemical Mass
Balance (CMB) receptor model.

Number of Sources/Species: Between 6 and 14 for each study.

#### Sources:

Varied with each study (see Results).

#### Methodology:

The article reviews in detail 22 different studies that used CMB analysis to determine the apportionment of  $PM_{2.5}$  and  $PM_{10}$ .

#### Other Analyses:

Varied with each study, including comparison to emission inventories, measurement of VOCs and gaseous pollutants, airshed models, dispersion modeling, wind trajectories, and back trajectories.

# **Findings and Recommendations**

#### Results:

For North American cities, total average  $PM_{10}$  mass ranged from 35 to 136  $\mu$ g/m<sup>3</sup> and for  $PM_{2.5}$  from 8 to 66  $\mu$ g/m<sup>3</sup>.

Most of the primary contributions to  $PM_{2.5}$  and  $PM_{10}$  were from diesel and gasoline powered vehicle exhaust. Primary contributions from "ducted" sources (e.g., power stations) were significant only when they did not have modern pollution controls. The contribution from secondary sulfates and nitrates was identified but could not be attributed to specific precursor sources using CMB or other chemical transport models. Use of source and receptor models together improved estimates of source contribution. While CMB was developed for primary pollutants, several studies adapted it to model secondary pollutants.

One concern with the use of CMB was the insufficient number of adequately characterized sources. This creates significant uncertainty in the results, which is not always correctly reflected in significant digits and correct standard errors. Some source profiles were developed with too few samples (e.g., diesel engine emissions), are out-of-date (e.g., many sources in the SPECIATE database), are not relevant to the current location (e.g., fuel constituents), or are not included in the correct proportions or at all (e.g., cooking, fugitive dust). Most studies did not adequately document their source profiles.

# Temporal Signals:

Not discussed in overall paper.

# Transboundary Transport:

Not discussed in overall paper.

<b>Study Reference:</b> Chow, J.C. and J.G. Watson (2002). Review of PM <sub>2.5</sub> and PM <sub>10</sub> apportionment for fossil fuel combustion and other sources by the chemical mass balance receptor model. <i>Energy &amp; Fuels</i> , <b>16</b> , 222-260. <b>Type of Study:</b> Literature survey <b>Funding Source:</b> None mentioned					
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations			
		Recommendations:  CMB could be used to verify and improve emission inventories and to apportion PM sources, both valuable for air quality policy and enforcement. However, much more work needs to be done to increase the sampling to adequately represent the chemical compositions of the sources.  Upcoming Studies/Planned Work:  None mentioned.			

**Study Reference:** Coutant, B., T. Kelly, J. Ma, B. Scott, B. Wood, and H. Main (2002). Source apportionment analysis of air quality monitoring data: Phase 1 Final Report. Prepared for the Mid-Atlantic/Northeast Visibility Union and Midwest Regional Planning Organization by Battelle Memorial Institute and Sonoma Technology, Inc., May 2002.

**Type of Study:** Contract research report

# **Site and Data Specifications**

# Site Location(s) and Type(s): 16 sites in Federal Class I and rural areas in the midwest, northeast, mid-Atlantic, and southeast plus one urban site (see Table 1 for complete list)

#### Timeframe:

Start date varied from 1988 to 1996 and end date from 1998 to 2000, depending on site.

#### Frequency:

Integrated 24-hour sampling taken 2 times per week.

#### Data Source:

10 IMPROVE sites and 6 CASTNET sites

#### Data Description:

Speciated PM<sub>2.5</sub> mass concentrations, total PM<sub>2.5</sub> mass concentration, and some total PM<sub>10</sub> mass concentrations.

Analysis methods included gravimetric, particle induced X-ray emission, proton elastic scattering analysis, and X-ray fluorescence. Additional meteorological data came from the EPA and the National Park

# **Source Apportionment Method**

Source Apportionment Tool(s):
PMF and UNMIX. PMF was used as
the primary tool and UNMIX was used
as an investigational tool and a
confirmation of PMF results.

# Number of Sources/Species:

9 sources per site for PMF and over 20 species

#### Sources:

Varied per site (see Results).

#### Methodology:

The data were sorted with a quality assurance and validation step to remove certain data measurements. Less than 10% of the data were removed. A check for long-term consistency was also conducted. This resulted in changing the timeframe for some of the datasets since certain discrete events (e.g., network shutdowns) influenced the data. Sources were determined using PMF with 22 inputs (total mass and sulfate, S, OC, EC, Na, Al, Ca, Si, Fe, K, Br, Cu, Mg, Mn, Ni, Pb, Se, Sr, Ti, V, and Zn). Certain metals were not modeled as well as others and it was noted that nitrate was so problematic to model it was not included as an input. After source identifications were

# $Findings\ and\ Recommendations$

**Funding Source:** U.S. EPA OAQPS

#### Results:

Total average PM<sub>2.5</sub> mass ranged from 8 to 18  $\mu$ g/m³. Sources interpreted as secondary sulfate and secondary organics were identified as major sources at nearly every site. Mass concentrations of secondary sulfates typically exceeded 4  $\mu$ g/m³ and may be attributed to large sulfur sources such as coal-fired power plants. Secondary organics typically exceeded 2  $\mu$ g/m³ and generally lacked tracers to attribute them to specific sources. In a few cases (Washington and Brigantine) additional species were able to identify the organic carbon as being from mobile sources.

Vegetative burning and/or wood smoke sources were found at most sites, particularly Jefferson/James River, Lye Brook, Mammoth Cave, and Shenandoah sites. Crustal soil dust was found at nearly all sites. Sources specific to certain sites were incinerator emissions (at 10 sites), magnesium source (Livonia, M.K. Goddard, and Quaker City), sea salt (Acadia, Brigantine, Shenandoah, and Washington DC), and other smaller sources.

When assessing the sources for the 20% best visibility days, no sources strongly dominated. For the 20% worst visibility dates, certain sources were dominant and in many cases over 50% of the light extinction could be attributed to secondary sulfate. For the IMPROVE sites (except Boundary Waters), the secondary sulfate source was 65-78% of the light extinction on the worst days. The secondary sulfate at the CASTNET sites was 41-65% on the worst days. For six sites, organic carbon was also strongly associated with the 20% worst days and peaks in the summer. The exception is Washington DC, where organic carbon peaks in the winter,

**Study Reference:** Coutant, B., T. Kelly, J. Ma, B. Scott, B. Wood, and H. Main (2002). Source apportionment analysis of air quality monitoring data: Phase 1 Final Report. Prepared for the Mid-Atlantic/Northeast Visibility Union and Midwest Regional Planning Organization by Battelle Memorial Institute and Sonoma Technology, Inc., May 2002.

Type of Study: Contract research report Funding Source: U.S. EPA OAQPS

Type of Study. Confract research report		runding bource: 0.5. El 17 On Q1 5
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Service Air Resources Division.	assigned, apportionments were done for the 20% best and 20% worst visibility days, since visibility is a key issue in the Class I areas.  Other Analyses: Light extinction was calculated using monthly relative humidity factors in order to determine the 20% best and 20% worst days. Also, speciate profiles were used as part of a verification process. The purpose was to check the consistency of the source profiles generated by the source apportionment modeling with speciate profiles where appropriate.	Ingeneral, the worst visibility is in the summer, best is in the winter. A site by site table presents a summary of seasonality of the sources. Most sources exhibited some seasonality.  Transboundary Transport: This study did not evaluate transport versus local sources.  Recommendations: Some possible data artifacts in the two datasets should be further investigated. The conceptual models need to be further refined to better model fine particle formation and transport. Temporal, spatial, and chemical gaps need to be filled in the ambient databases. Emissions inventories and back trajectory models could be used in conjunction with the source apportionment results to better understand sources.  While source profiles were not used in the model, they were compared with the results, which led to several recommendations. The source profiles need to be improved, including systematic sampling to obtain profiles for specific sources, evaluating data to better understand rural and urban profiles, and refining methods to compare measurement and model results. For example, further analysis is needed to determine the sources of organic carbon as well as how the source can be better identified. The various receptor techniques should be compared. Case studies of certain source types should be conducted to aid characterization.  Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Coutant, B.W., C.H. Holloman, K.E. Swinton, and H.R. Hafner. (2003). "Eight-site source apportionment of PM<sub>2.5</sub> speciation trends data." Revised Draft Report for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Contract No. 68-D-02-061, Work Assignment No. 1-05, April 30, 2003.

**Type of Study:** Contract research report

# **Site and Data Specifications**

# Site Location(s) and Type(s): Bronx, NY (urban site); St. Louis, MO (urban site); Houston, TX (urban site); Washington, DC (urban site); Milwaukee, WI (urban site); Birmingham, AL (urban site); Charlotte, NC (urban site); Indianapolis, IN (urban site)

#### Timeframe:

 $\sim$ 160 days each site in 2001 and 2002

#### Frequency:

Integrated 24 hour samples taken every third day.

#### Data Source:

EPA PM<sub>2.5</sub> speciation trends sites, supplemented by data from the New York Department of Environmental Conservation (for Bronx site)

#### Data Description:

Total PM<sub>2.5</sub> and PM<sub>2.5</sub> speciation data. Analytical methods included X-ray fluorescence, ion chromatography, and thermal-optical analysis. Data

# Source Apportionment Method

Source Apportionment Tool: PMF for primary analysis, UNMIX for preliminary analysis

Number of Sources/Species: 6-8 sources per site and 28 species

#### Sources:

Varied across the 3 sites (see Results)

#### Methodology:

Study used receptor modeling and careful data quality checks, including review for internal consistency and interviews with local monitoring personnel. Sources were determined with PMF (including total PM<sub>2.5</sub> and sulfate, nitrate, ammonium, Al, As, Ba, Br, Ca, Cl, Cr, Cu, EC, Fe, Pb, Mn, Ni, OC, K, K<sup>+</sup>, Se, Si, Na, S, Sn, Ta, Ti, V, and Zn).

# Other Analyses:

The output of the source apportionment was combined with back trajectories to identify source locations 3-72 hours upwind. Pollution and wind roses were also used to help identify source directions.

# Findings and Recommendations

**Funding Source:** U.S. EPA OAQPS

#### Results:

Bronx, NY: Mean mass= $16.1 \,\mu\text{g/m}^3$ . Identified sources (in decreasing order) were: ammonium sulfate (5.29  $\mu\text{g/m}^3$ , transport from PA, VA, MA, WV), ammonium nitrate (4.09  $\mu\text{g/m}^3$ ), mobile-tire wear combination (2.49  $\mu\text{g/m}^3$ , major highways), industrial (1.82  $\mu\text{g/m}^3$ , local & transport), fuel oil (1.22  $\mu\text{g/m}^3$ , local & non-local oil-fired power plants), crustal (0.97  $\mu\text{g/m}^3$ , street cleaning, farming, mining), and sea spray (0.30  $\mu\text{g/m}^3$ ).

St. Louis, MO: Mean mass=17.2  $\mu$ g/m³. Identified sources were: coal combustion (5.74  $\mu$ g/m³, known sources east of site), ammonium nitrate (5.02  $\mu$ g/m³, power plants), mobile (2.92  $\mu$ g/m³, local highways, high in lead from road dust), crustal (1.43  $\mu$ g/m³, agricultural or industrial), zinc refinery (0.85  $\mu$ g/m³, local industry), steel production (0.76  $\mu$ g/m³, local industry), and copper smelting (0.59  $\mu$ g/m³, local industry).

Houston, TX: Mean mass=14.2  $\mu$ g/m³. Identified sources were: ammonium sulfate from fossil fuels (5.54  $\mu$ g/m³, local), mobile sources (5.19  $\mu$ g/m³, local), mobile-Mn (1.04  $\mu$ g/m³, local, ship channel), industrial (0.87  $\mu$ g/m³, local), crustal (0.77  $\mu$ g/m³, local), fireworks (0.49  $\mu$ g/m³, one-time local, with some wood smoke), and marine-nitrate (0.29  $\mu$ g/m³, local sea-breeze).

Washington, DC: Mean mass= $16.7 \,\mu\text{g/m}^3$ . Identified sources were: ammonium sulfate (7.70  $\mu\text{g/m}^3$ ), mobile sources (4.72  $\mu\text{g/m}^3$ ), road dust and diesel (1.47  $\mu\text{g/m}^3$ , possibly from a road construction project), ammonium nitrate and salt (1.23  $\mu\text{g/m}^3$ ), Canadian fires (1.11  $\mu\text{g/m}^3$ ), and fireworks (0.53  $\mu\text{g/m}^3$ , with

**Study Reference:** Coutant, B.W., C.H. Holloman, K.E. Swinton, and H.R. Hafner. (2003). "Eight-site source apportionment of PM<sub>2.5</sub> speciation trends data." Revised Draft Report for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Contract No. 68-D-02-061, Work Assignment No. 1-05, April 30, 2003.

Type of Study: Contract research report Funding Source: U.S. EPA OAQPS

Type of Study: Contract research report		runding source: U.S. EFA OAQFS
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
screening was designed to retain		some vegetative burning).
trace metal results.		Milwaukee, WI: Mean mass=14.5 $\mu$ g/m³. Identified sources were: ammonium sulfate (4.54 $\mu$ g/m³), ammonium nitrate (4.07 $\mu$ g/m³), Cl source (2.66 $\mu$ g/m³, unknown), mobile sources (1.53 $\mu$ g/m³), diesel based sulfate (0.93 $\mu$ g/m³, unclear), fireworks (0.36 $\mu$ g/m³, with some vegetative burning), crustal related events (0.19 $\mu$ g/m³), and crustal (0.12 $\mu$ g/m³).
		Birmingham, AL: Mean mass=19.5 $\mu$ g/m³. Identified sources were: secondary sulfate (7.27 $\mu$ g/m³, regional transport), mobile sources (6.51 $\mu$ g/m³), ammonium nitrate (1.84 $\mu$ g/m³, coal), crustal (1.27 $\mu$ g/m³), fireworks (1.15 $\mu$ g/m³, with some vegetative burning), Zn source (0.79 $\mu$ g/m³, industrial), and lead source (0.71 $\mu$ g/m³, single event).
		Charlotte, NC: Mean mass= $16.2\mu g/m^3$ . Identified sources were: secondary sulfate (5.71 $\mu g/m^3$ , coal), mobile (3.87 $\mu g/m^3$ ), oil combustion (1.87 $\mu g/m^3$ ), nitrate (1.21 $\mu g/m^3$ ), smelting (0.67 $\mu g/m^3$ ), crustal (0.57 $\mu g/m^3$ ), fireworks (0.48 $\mu g/m^3$ , with some vegetative burning), and sea salt (0.08 $\mu g/m^3$ ).
		Indianapolis, IN: Mean mass=17.3 $\mu$ g/m³. Identified sources were: ammonium sulfate (7.03 $\mu$ g/m³), ammonium nitrate (3.58 $\mu$ g/m³), mobile sources (3.21 $\mu$ g/m³), winter coal (1.64 $\mu$ g/m³), fireworks (0.69 $\mu$ g/m³, with some vegetative burning), crustal (0.51 $\mu$ g/m³), sea salt (0.47 $\mu$ g/m³, long range transport), and Canadian fires (0.25 $\mu$ g/m³).
		Overall, sulfate, from coal or oil based sources, accounted for about one-third of the PM <sub>2.5</sub> . Nitrates and mobile sources are the other largest portions depending on meteorological

**Study Reference:** Coutant, B.W., C.H. Holloman, K.E. Swinton, and H.R. Hafner. (2003). "Eight-site source apportionment of PM<sub>2.5</sub> speciation trends data." Revised Draft Report for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Contract No. 68-D-02-061, Work Assignment No. 1-05, April 30, 2003.

**Type of Study:** Contract research report

Funding Source: U.S. EPA OAQPS

Type of Study: Contract research report		Funding Source: U.S. EPA OAQPS
<b>Site and Data Specifications</b>	<b>Source Apportionment Method</b>	Findings and Recommendations
		conditions. Crustal sources appear at all sites at low levels. The other sources are site specific.
		Temporal Signals: Temporal signals varied with each site. In general, the sulfate source was higher in the summer and the nitrate source higher in the winter. Mobile sources varied but there were some higher values in the fall. The other sources were more uniform, with the exception of the events (e.g., fireworks, Canadian fires).
		Transboundary Transport: Secondary sulfate/coal combustion sources generally corresponded to utility plants in the midwest or oil combustion from the south. Intense smoke plumes from fires in Canada in early July influenced several sites.
		Recommendations: Combination of techniques provided useful method to understanding PM <sub>2.5</sub> at urban sites, but sufficient data must be available, back trajectory methods must be carefully validated, and wind/pollution roses must be used carefully since they are highly variable and localized. A "weight-of-evidence" approach may be required.
		Upcoming Studies/Planned Work: None mentioned

Study Reference: Kim, E., and P.K. Hopke (2003). "Source Apportionment of Fine Particles at Washington, DC utilizing Temperature Resolved Carbon Fractions." Submitted to *Journal of the Air Waste Management Association*.

Type of Study: Peer reviewed records

Type of Study: Peer reviewed research		Funding Source: U.S. EPA	
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations	
	Source Apportionment Method  Source Apportionment Tool (s):    PMF  Number of Sources/Species:    10 sources and 35 species  Sources:    Sulfate-rich secondary aerosol I, gasoline vehicle, sulfate-rich secondary aerosol II, nitrate-rich secondary aerosol, sulfate-rich secondary aerosol, sulfate-rich secondary aerosol III, incinerator, aged sea salt, airborne soil, diesel emissions, and oil combustion.  Methodology:    In addition to the standard speciation variables, this study used 8 carbon fractions to separate gasoline and diesel vehicle emissions.  Concentration values were used for measured data and values below the detection limit were replaced by half the	Findings and Recommendations  Results:  The arithmetic mean of the total mass was 17.9 μg/m³. The sources identified, in percent mass concentration, were:  Sulfate-rich secondary aerosol I (43%), gasoline vehicle (21%), sulfate-rich secondary aerosol II (11%), nitrate-rich secondary aerosol (9%), sulfate-rich secondary aerosol III (6%), incinerator (4%), aged sea salt (2%), airborne soil (2%), diesel emissions (2%), and oil combustion (2%).  The study compares its results to Song et al (2001), which was a similar analysis but with 2 carbon fractions instead of 8.  This study split the motor vehicle source into gasoline and diesel; their total (23.8%) was significantly higher than the 9.0% in Song et al (2001). In this study, diesel emissions may include contributions from metal processing sources, based on evidence of high metals in the source. Part of the mass attributed to gasoline vehicles in this study had been previously assigned to OC associated with secondary nitrate. The gasoline source has high OC3 and OC4 and diesel has high EC. The CPF plot for the gasoline vehicle source points to a nearby highway.  Temporal Signals:	
	measured data and values below the	to a nearby highway.	
	variously set, depending on the data (measured, below detection limit, missing). Also, the estimated uncertainties of Al and NO <sub>3</sub> were increased by factors of 2 and 3, respectively, to decrease their weight in	the weekends. Oil combustion has a winter peak.  Transboundary Transport: HYSPLIT on a peak airborne soil day indicated influence from a Saharan dust storm on July 7, 1993. The oil combustion source likely comes from the northeast urban corridor.	

Study Reference: Kim, E., and P.K. Hopke (2003). "Source Apportionment of Fine Particles at Washington, DC utilizing Temperature Resolved Carbon Fractions." Submitted to Journal of the Air Waste Management Association.

Type of Study: Peer reviewed research		Funding Source: U.S. EPA	
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations	
	the model fit. The uncertainties of OC1 and EC1 were increased by a factor of 2 to represent known uncertainties. PMF was used with 35 input species, not including total mass (Al, As, Br, Ca, Cl, Cr, Cu, Fe, H, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Se, Si, Sr, Ti, V, Zn, Zr, OC1, OC2, OC3, OC4, OP, EC1, EC2, EC3, SO4, and NO3). Total mass was apportioned by regression. The robust mode was used. The number of sources, FPEAK, FKEY, and initial values were varied to obtain final solution.  Other Analyses:  The conditional probability function (CPF) was used to analyze point source impacts from various wind directions. CPF is a ratio of the number of occurrences from a wind sector divided by the total number of data from that wind sector. HYSPLIT was used to calculate air mass back trajectories for high impact days.	Recommendations: Temperature resolved carbon fraction data can be used to separate gasoline and diesel sources, as well as improve source apportionment results.  Upcoming Studies/Planned Work: None mentioned.	

Study Reference: Kim, E., P.K. Hopke, and E.S. Edgerton (2003a). "Improving Source Identification of Atlanta Aerosol using Temperature Resolved Carbon Fractions in Positive Matrix Factorization" Submitted to Aerosol Science and Technology

Type of Study: Peer reviewed rese		Funding Source: Southern Company
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): Atlanta (urban)	Source Apportionment Tool (s): PMF	Results: The arithmetic mean of the total mass was $18.3 \ \mu g/m^3$ . The
Timeframe: 1998-2000	Number of Sources/Species: 11 sources and 28 species.	sources identified, in percent mass concentration, were: sulfate-rich secondary aerosol I (50%), on-road diesel emissions (11%), nitrate-rich secondary aerosol (9%), wood
Frequency: Daily integrated 24-hour samples.	Sources: Sulfate-rich secondary aerosol I, onroad diesel emissions, nitrate-rich	smoke (7%), gasoline vehicle (6%), sulfate-rich secondary aerosol II (6%), metal processing (3%), airborne soil (3%), railroad traffic (3%), cement kiln/carbon-rich (2%), and bus
Data Source: Southeastern Aerosol Research and Characterization (SEARCH) air monitoring network.  Data Description: Speciated PM <sub>2.5</sub> data. Analyzed with energy dispersive X-ray fluorescence, ion chromatography, and IMPROVE/thermal optical reflectance protocol for carbon	secondary aerosol, wood smoke, gasoline vehicle, sulfate-rich secondary aerosol II, metal processing, airborne soil, railroad traffic, cement kiln/carbon-rich, and bus maintenance facility/highway traffic.  Methodology: In addition to the standard speciation variables, this study used carbon fractions to separate gasoline and diesel vehicle emissions.  Concentration values were used for	maintenance facility/highway traffic (2%).  This study is comparable to Kim et al (2002a) and Kim et al (2003b), which used only the traditional OC and EC data, not the 8 carbon fractions. Using the 8 carbon fractions, this study identified four traffic-related combustion sources (gasoline vehicle, on-road diesel, railroad, and bus maintenance facility) with carbon fractions whose abundances were different between the various sources. The gasoline vehicle source was high in OC3 and OC4, on road diesel was high in EC1 and OC2, railroad source was high in EC2, and bus maintenance/highway traffic was high in EC1 and OC3 (a bus maintenance facility was about 200 m southeast of the site)
fractions.	measured data and values below the detection limit were replaced by half the detection limit. Missing values were replaced by the geometric mean of the measured values. Uncertainties were variously set, depending on the data (measured, below detection limit, missing). Also, the estimated	Temporal Signals: The sulfate-rich secondary aerosol I source shows strong seasonal variation with higher concentrations in the summer. Sulfate-rich secondary aerosol II was slightly higher in the summer. The nitrate-rich secondary aerosol source had a maxima in the winter and there was a winter increase from metal recycling.  . Wood smoke source was high in the winter (residential wood)
	uncertainties of certain elements were increased to decrease their weight in the	burning) with short peaks in spring and summer, likely due to

increased to decrease their weight in the

forest fires and prescribed burning, which was confirmed by

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	model fit: EC3, As, Se, and Sn (by 2x); Cl, Mn, Pb, Sb, Ti, Al, Fe (3x); and Br (4x). Also, due to inadequate Se data, the estimated uncertainties of NH <sub>4</sub> <sup>+</sup> and SO <sub>4</sub> <sup>=</sup> were increased by a factor of 4 to decrease their weight in the model fit. The uncertainties of OC1 and EC1 were increased by a factor of 4 to represent known uncertainties.  PMF was used with 28 input species (As, Br, Cu, Mn, Pb, Sb, Se, Sn, Ti, Zn, Al, Si, K, Ca, Fe, Cl, OC1, OC2, OC3, OC4, OP, EC1, EC2, EC3, SO <sub>4</sub> , NO <sub>3</sub> , NH <sub>4</sub> and PM <sub>2.5</sub> ). Total mass was included as an independent variable in the PMF modeling and its uncertainty set to 4 times its value to decrease its weight in the model.  The robust mode was used. The final solution was determined running PMF with different FPEAK and FKEY values to determine an optimal solution.  Other Analyses: The conditional probability function (CPF) was used to analyze point source impacts from various wind directions. CPF is a ratio of the number of occurrences from a wind sector divided by the total number of data from that wind sector.	the CPF plots. Airborne soil is higher in the summer, likely due to road dust resuspension.  The bus maintenance/highway traffic source was higher on the weekdays (the facility only operated on weekdays). All traffic related sources contributed more to the total mass in the winter, except for railroad traffic which was higher in the summer.  Transboundary Transport: None mentioned.  Recommendations: Temperature resolved carbon fraction data can be used to separate gasoline and diesel sources, as well as improve source apportionment results.  Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Kim, E., P.K. Hopke, and E.S. Edgerton (2003b). "Utilizing Hourly Wind Measurements as Independent Variables in Multilinear Receptor Model Studies of Atlanta aerosol." Submitted to *Atmospheric Environment*.

**Type of Study:** Peer reviewed research **Funding Source:** Southern Company

Multilinear Receptor Model Studie <b>Type of Study:</b> Peer reviewed rese	
Site and Data Specifications	
Site Location(s) and Type(s): Atlanta, GA (urban)	
Timeframe: 1998-2000	
Frequency: Daily integrated 24-hour samples.	
Data Source: Southeastern Aerosol Research and Characterization (SEARCH) air monitoring network.	
Data Description: Speciated PM <sub>2.5</sub> data. Analyzed with energy dispersive X-ray	

Speciated PM<sub>2.5</sub> data. Analyzed with energy dispersive X-ray fluorescence, ion chromatography, and IMPROVE/thermal optical reflectance protocol for carbon fractions. Wind speed and direction were measured hourly.

# **Source Apportionment Method**

Source Apportionment Tool (s): Multilinear Engine (ME)

# Number of Sources/Species:

9 sources and 22 elemental species, 2 wind variables, and three time variables.

#### Sources:

Sulfate-rich secondary aerosol I, gasoline vehicle, diesel emissions, nitrate-rich secondary aerosol, metal processing, wood smoke, airborne soil, sulfate-rich secondary aerosol II, and the mixture of cement kiln with a carbon-rich source.

# Methodology:

Concentration values were used for measured data and values below the detection limit were replaced by half the detection limit. Missing values were replaced by the geometric mean of the measured values. Uncertainties were variously set, depending on the data (measured, below detection limit, missing). Also, the estimated uncertainties of certain elements were increased to decrease their weight in the model fit: PM<sub>2.5</sub> mass concentration, As, Se, Ti, Si, K, and Ca (by 2x); OC, EC, Cl, Mn, Sb, Sn, and Al (3x); SO<sub>4</sub><sup>=</sup>, NH<sub>4</sub><sup>+</sup>, Br, and Fe (4x); and Cu (5x).

# **Findings and Recommendations**

#### Results:

The arithmetic mean of the total mass was  $18.0 \,\mu\text{g/m}^3$ . The sources identified, in percent mass concentration, were: sulfate-rich secondary aerosol I (54%), gasoline vehicle (15%), diesel emissions (11%), nitrate-rich secondary aerosol (9%), metal processing (3%), wood smoke (3%), airborne soil (2%), sulfate-rich secondary aerosol II (2%), and the mixture of cement kiln with a carbon-rich source (0.9 %).

Compared to a similar study using PMF (Kim et al 2002a), ME was able to separate two sulfate-rich secondary aerosols and two mobile sources (gasoline and diesel vehicles). The other sources were similar between the studies, although their percent contributions did vary, likely due to strong directionality of some sources.

# Temporal Signals:

For the nitrate-rich secondary aerosol, values decreased with increasing wind speed, indicating dilution. The other sources did not show a clear trend with wind speed.

The nitrate-rich secondary aerosol was also higher in the morning and decreased in the afternoon, likely due to meteorological effects. The other sources did not show strong diurnal effects.

The sulfate-rich secondary aerosol I source showed strong seasonal variation with a maxima in the summer. The sulfate-rich secondary aerosol II source (with a high OC concentration) was slightly higher in the winter. Nitrate-rich secondary aerosols were also higher in the winter.

Other variations included low gasoline and higher diesel contributions in the fall, metal processing and wood smoke

Study Reference: Kim, E., P.K. Hopke, and E.S. Edgerton (2003b). "Utilizing Hourly Wind Measurements as Independent Variables in Multilinear Receptor Model Studies of Atlanta aerosol." Submitted to <i>Atmospheric Environment</i> .  Type of Study: Peer reviewed research  Funding Source: Southern Company		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	ME was used with 21 input species (As, Br, Cu, Mn, Pb, Sb, Se, Sn, Ti, Zn, Al, Si, K, Ca, Fe, Cl, EC, OC, SO <sub>4</sub> , NO <sub>3</sub> , NH <sub>4</sub> and PM <sub>2.5</sub> ). Total mass was included as an independent variable in the ME model and its uncertainty set to 4 times its value to decrease its weight in the model. Also, wind direction (hourly average), wind speed (hourly average), time of day, time of year, and weekend/weekday variables were used as variables in ME.  The robust mode was used. The final solution was determined by running ME hundreds of times varying the number of sources and uncertainty estimates to determine an optimal solution.  Other Analyses: None.	higher in the winter, and airborne soil and cement kiln higher in the spring and summer.  Diesel emission and cement kiln emission were lower on the weekends.  Transboundary Transport: None mentioned.  Recommendations: The results of this study indicate that time resolved meteorological measurements helps separate diesel emissions from gasoline vehicle emissions and could significantly improve source apportionment studies.  Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Kim, E., P.K. Hopke, and E.S. Edgerton (2002a). "Source Identification of Atlanta Aerosol by Positive Matrix Factorization." Submitted to *Journal of the Air Waste Management Association*.

Type of Study: Peer reviewed research		Funding Source: Southern Company	
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations	
Site and Data Specifications  Site Location(s) and Type(s): Atlanta, GA (urban)  Timeframe: 1998-2000  Frequency: Daily integrated 24-hour samples.  Data Source: Southeastern Aerosol Research and Characterization (SEARCH) air monitoring network.  Data Description: Speciated PM <sub>2.5</sub> data. Analyzed with energy dispersive X-ray fluorescence, ion chromatography, and	Source Apportionment Method  Source Apportionment Tool (s):    PMF  Number of Sources/Species:    Fine PM: 8 sources and 25 species    Coarse PM. 5 sources and 15 species  Sources:    Varied between PM fine and PM coarse    (see Results).  Methodology:    Separate analysis was conducted for    fine particles and coarse particles. For    fine particles, the nitrate mass from the    nylon filter of the 3-stage filter pack    was added to the measured particle    mass from the Teflon filter in order to    resolve some of the differences between    the sum of the species mass and the	Findings and Recommendations  Results:  The arithmetic mean of the total fine mass was 18.0 μg/m³. The sources identified, in percent mass concentration, were: sulfate-rich secondary aerosol (56%), motor vehicle (22%), wood smoke (11%), nitrate-rich secondary aerosol (7%), mixed source of cement kiln and organic carbon (2%), airborne soil (1%), metal recycling facility (0.5%), and mixed source of bus station and metal processing (0.3%).  The motor vehicle source, high in OC, EC, and soil dust constituents (Si, Fe), was a combination of gasoline and diesel sources, as well as resuspended road dust.  The arithmetic mean of the total coarse mass was 10.1 μg/m³. The sources identified, in percent mass concentration, were: airborne soil (60%), nitrate-rich secondary aerosol (16%), sulfate-rich secondary aerosol (12%), cement kiln (11%), and metal recycling facility (1%).  Temporal Signals:	
chromatography, and IMPROVE/thermal optical reflectance protocol for carbon fractions.	particle mass.	Fine particles: The sulfate-rich secondary aerosol, including NH <sub>4</sub> <sup>+</sup> as 30% of its mass, was high in the summer. The mixed source and airborne soil were also higher in the summer. Wood smoke source was high in the winter (residential wood burning) with short peaks in spring and summer, likely due to forest fires and prescribed burning. The nitrate-rich secondary aerosol had a maxima in the winter, and there were slight winter increases in metal recycling and motor vehicles.  Coarse particles: None had strong temporal trends, although there was a slight increase the metal recycling source in the summer and in the cement kiln sources in the winter.  Transboundary Transport:	

Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
	factor of 4 to decrease their weight in the model fit.  For fine PM, PMF was used with 25 input species (As, Ba, Br, Cu, Mn, Pb, Sb, Se, Sn, Ti, Zn, Cr, Ni, V, Al, Si, K, Ca, Fe, Cl, OC, EC, SO <sub>4</sub> , NO <sub>3</sub> , NH <sub>4</sub> and PM <sub>2.5</sub> ). For coarse PM, PMF was used with 15 species (Cr, Cu, Fe, Mn, Ni, V, Al, Si, K, Ca, Ti, SO <sub>4</sub> , NO <sub>3</sub> , NH <sub>4</sub> and PM <sub>coarse</sub> ). Total mass was included as an independent variable in the PMF modeling and its uncertainty set to 4 times its value to decrease its weight in the model.  The robust mode was used. The final solution was determined running PMF with different FPEAK values to determine an optimal solution.  Other Analyses:  The conditional probability function (CPF) was used to analyze point source impacts from various wind directions. CPF is a ratio of the number of occurrences from a wind sector divided by the total number of data from that wind sector.	None mentioned.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.

	' Submitted to Environmental Science & Tech earch	"Analysis of Ambient Particle Size Distributions using UNMIX Innology.  Funding Source: University of Washington/EPA Northwest Research Center for Particulate Air Pollution and Health, and by University of Rochester/EPA Particulate Matter and Health Center
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Seattle, WA (urban)	Source Apportionment Tool: UNMIX and PMF	Results: Both PMF and UNMIX identified four similar underlying
Timeframe: Dec 2000-Feb 2001	Number of Sources/Species: 4 sources and 16 size intervals	factors in the size distributions. Factor 1 was an accumulation mode particle distribution with highest concentration between 0.2 and 0.3 µm and nocturnal pattern, particularly on
Frequency: Hourly	Sources: Wood burning, secondary aerosol,	weekends. CPF indicated a source location in nearby residential areas. This factor was likely from residential wood
Data Source: University of Washington, PM Center Data Description:	diesel emissions, and motor vehicle emissions  Methodology: Study used size distribution data with	burning.  Factor 2 was a larger particle distribution with a noon and evening high pattern from the direction of the ocean and nearby highways. This was likely secondary aerosol.
PM diameter distribution data collected with a differential mobility particle sizer (DMPS) with 16 size intervals (20-400 nm). Also, used NO <sub>X</sub> , CO, b <sub>sp</sub> , and b <sub>ap</sub> .  Size distribution profiles. First, use UNMIX to extract four factor extracted 4 factors in PMF to with UNMIX results.  Size distribution data without concentrations and below detervalues were not used.  PMF was run several times with different FPEAK values to obsoptimal solution. The robust sused. PMF results were norm with hourly total volume concentration were included as	UNMIX and PMF to compare to size distribution profiles. First, used UNMIX to extract four factors. Then, extracted 4 factors in PMF to compare with UNMIX results.	Factor 3 peaked around 0.1 $\mu$ m with a regular weekday-high rush hour pattern and correlation with NO <sub>X</sub> , CO, and b <sub>ap</sub> . The particle size distribution and the other factors indicated diesel related emissions.
		Factor 4 had a bimodal particle size distribution (peaks at 0.04 and 0.3 µm), a regular rush hour pattern on weekdays and weekends, and an association with NO <sub>x</sub> . CPF indicated a source location near local highways. This suggested a motor
	PMF was run several times with different FPEAK values to obtain the optimal solution. The robust mode was used. PMF results were normalized with hourly total volume concentrations. In UNMIX, the sum of volume concentration were included as an input. Results from both models were	vehicle source.  Percent contribution results were as follows (% from PMF / % from UNMIX): wood burning (48% / 44%), secondary aerosol (21% / 16%), diesel emissions (20% / 30%), and gas/vehicle combination (11% / 10%). Error analysis indicated the only significant difference between the PMF and UNMIX results was for Factor 3 (diesel emissions) where UNMIX estimated

<b>Study Reference:</b> Kim, E., P.K. Hopke, T.V. Larson, and D.S. Covert (2002b). and Positive Matrix Factorization." Submitted to <i>Environmental Science &amp; Tech</i> <b>Type of Study:</b> Peer reviewed research		•
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	correlated with NO <sub>X</sub> , CO, b <sub>sp</sub> , and b <sub>ap</sub> .	higher contribution than PMF.
	Other Analyses: The conditional probability function (CPF) was used to analyze point source impacts from various wind directions. CPF is a ratio of the number of occurrences from a wind sector divided by the total number of data from that wind sector.	Temporal Signals: Wood burning was higher on weekdays and in the evenings. The two mobile sources also exhibited daily a rush hour pattern.  Transboundary Transport: None mentioned.  Recommendations: Hourly size distribution data can be used with UNMIX and PMF. Speciation data would be needed to confirm the source identifications,  Upcoming Studies/Planned Work: Plan to use chemical speciation data at the same site and time periods to verify source identification.

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Spokane, WA (residential)  Timeframe: 1995-1997  Frequency: Daily integrated 24-hour samples.  Data Source: Special study  Data Description: Speciated PM <sub>2.5</sub> data. Analyzed with energy dispersive X-ray fluorescence, thermal manganese oxidation method, and ion chromatography. Also used hourly NO <sub>X</sub> and CO.	Source Apportionment Tool (s):     PMF  Number of Sources/Species:     7 sources and 16 species  Sources:     Vegetative burning, sulfate aerosol, motor, vehicle, nitrate aerosol, airborne soil, chlorine-rich source, and metal processing.  Methodology:     Concentration values were used for measured data and values below the detection limit were replaced by half the detection limit. Missing values were replaced by the geometric mean of the measured values. Uncertainties were variously set, depending on the data (measured, below detection limit, missing). Also, the estimated uncertainties of certain elements were increased to decrease their weight in the model fit: total C and Br (by 2x); Al and NH <sub>4</sub> <sup>+</sup> (3x); and Pb (4x).  PMF was used with 28 input species (As, Br, Cu, Mn, Pb, Zn, Al, Si, K, Ca,	Results: The arithmetic mean of the total fine mass was 12.1 µg/m³. The sources identified, in percent mass concentration, were: vegetative burning (44%), sulfate aerosol (19%), motor, vehicle (11%), nitrate aerosol (9%), airborne soil (9%), chlorine-rich source (possibly from a medical incinerator) (6%), and metal processing (3%).  Temporal Signals: Vegetative burning has a winter peak (residential fireplaces and wood stoves), summer minimum, with short-term peaks in late summer (forest fires and grass burning) and early fall (burning of agricultural field stubble).  Nitrate aerosols had a strong winter maxima due to lower temperature and higher humidity in the winter.  Motor vehicle sources had a slight winter high and airborne soil was higher in the summer. Both the chlorine-rich and metal processing sources had sharp peaks without a seasonal pattern.  Transboundary Transport: None mentioned.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.

<b>Study Reference:</b> Kim, E., T.V. Larson, P.K. Hopke, C. Slaughter, L.E. Sheppard, and C. Claiborn (2002c). "Source Identification of PM2.5 in an arid Northwest U.S. City by Positive Matrix Factorization." Submitted to <i>Atmospheric Research</i> .		
Type of Study: Peer reviewed re		Funding Source: Mickey Leland National Urban Air Toxics
		Research Center, University of Washington/EPA Northwest
		Research Center for Particulate Air Pollution and Health, and by
		University of Rochester/EPA Particulate Matter and Health
		Center
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	Fe, Cl, C, SO <sub>4</sub> , NO <sub>3</sub> , and NH <sub>4</sub> ). Total mass was apportioned by regression. The robust mode was used. The FPEAK, FKEY, and number of sources were varied to obtain the optimal solution.  Other Analyses: The conditional probability function (CPF) was used to analyze point source impacts from various wind directions. CPF is a ratio of the number of occurrences from a wind sector divided by the total number of data from that wind sector.	

Study Reference: Lee, J.H, Y. Yoshida, B. Turpin, P.K. Hopke, R.L. Poirot, P.J. Lioy and J.C. Oxley (2003a). Identification of sources contributing to the Mid-Atlantic regional aerosol; submitted to Journal of the Air and Waste Management Association.

**Type of Study:** Peer reviewed research Funding Source: New Jersey Department of Environmental

Type of Study. Feel leviewed research		Protection and New Jersey Agricultural Experiment Station
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): Brigantine National Wildlife Refuge, NJ (Class I visibility area)  Timeframe: 1991-1999  Frequency: Integrated 24-hour samples every Wednesday and Saturday  Data Source: IMPROVE network  Data Description: Speciated PM <sub>2.5</sub> data. Analytical techniques included gravimetric, X-ray fluorescence, proton elastic scattering analysis, proton induced X-ray emission, ion chromatography, and thermal-optical reflectance.	Source Apportionment Method  Source Apportionment Tool (s):    PMF  Number of Sources/Species:    9 sources and 30 species.  Sources:    Summer coal combustion, winter coal combustion, motor vehicle/mixed combustion, diesel/Zn-Pb, incinerator/industrial, oil combustion, soil, aged sea salt, and fresh sea salt.  Methodology:    Concentrations and sources determined with PMF and confirmed with two types of trajectory analysis. Key assumptions were that all elemental S is sulfate and all sulfate is ammonium sulfate, which might overestimate sulfate mass. Also, assumed all nitrate is ammonium nitrate, which may also	Results:  Average PM <sub>2.5</sub> mass = 11.4 μg/m³, with a summer average of 15.6 μg/m³ and the other seasons approximately 10 μg/m³. The three most abundant species in the annual fine particulate mass were sulfate (49% with mean concentration of 5.6 μg/m³), organic compounds (22%, 2.5 μg/m³), and ammonium nitrate (10%). For sources, coal combustion in both summer and winter were 66% of the total fine mass concentration with the remaining 8 sources under 10% each. Local sources of oil combustion, motor vehicle/mixed, and waste incinerator/ industrial contributed 17% and surround the site.  The oil combustion trajectories were usually slow moving air masses that passed through major east coast cities. Coal combustion was primarily from west and southwest of the site, with highest concentrations from fast moving air masses from the midwest. Diesel/Zn-Pb was north and west of the site.  Two types of sea salt factors were identified, including an aged sea salt possibly from sea breeze conditions.  Temporal Signals:
and thermal-optical reflectance.	overestimate nitrate. Soil versus non-soil (smoke) K was estimated by assuming the ratio of K/Fe in the soil was 0.6. PMF was used with 30 input species, not including total mass (As, Br, Ca, Cl, Cl, Cr, Cu, Fe, H, K, Mg,	For the species, sulfate concentration is highest in the summer and lowest in the winter. Organic compounds had higher concentrations in the summer but contribute more in the winter. Nitrate and "smoke K" were higher in the winter. Fine soil dust was higher in the summer.
	Mn, Mo, Na, Ni, nitrate, P, Pb, Rb, S, Se, Al, Si, Sr, Ti, V, Zn, Zr, OC, and EC). The PMF analysis was conducted in both the robust and non-robust modes	For sources, the summer coal combustion peaked in the summer, and winter coal combustion, oil combustion, motor vehicle/mixed combustion, and diesel/Zn-Pb peaked in the winter. Aged sea salt was higher in the spring, soil was higher

Study Reference: Lee, J.H, Y. Yoshida, B. Turpin, P.K. Hopke, R.L. Poirot, P.J. Lioy and J.C. Oxley (2003a). Identification of sour contributing to the Mid-Atlantic regional aerosol; submitted to <i>Journal of the Air and Waste Management Association</i> .  Type of Study: Peer reviewed research  Funding Source: New Jersey Department of Environment of Envi		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	and the results compared; the differences between the results were not significant.  Other Analyses: HYSPLIT trajectory analysis.  Cluster analysis in SAS/STAT version 6.12 for backward air trajectories on low and high concentration days.	in the summer, and fresh sea salt did not show a seasonal variation. Oil combustion was declining over the 1991-1999 sampling period reflecting a shift toward natural gas and warm winters in 1997 and 1998.  Transboundary Transport: Three dates (29 June 1994, 9 July 1994, and 26 August 1998) were identified as possibly influenced by Saharan dust by both the ratio of Al/Ca and back trajectories  Recommendations: Brigantine is a good location to study regional aerosols in a remote location; thus, results such as these could be used in the development of SIPs in the Mid-Atlantic.  Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Lee, P.K.H., J.R. Brook, E. Dabek-Zlotorzynska, and S.A. Mabury (2003b). Identification of the major sources contributing to PM<sub>2.5</sub> observed in Toronto. Submitted to *Environmental Science & Technology*.

Type of Study: Peer reviewed research

Funding Source

**Funding Source:** Toxic Substances Research Initiative, Government of Canada

including U.S. Midwest and Canadian power plants.

Recommendations:

		Government of Canada	
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations	
Site Location(s) and Type(s): Toronto, ON (urban)	Source Apportionment Tool: PMF	Results: Median PM <sub>2.5</sub> mass = $12.7 \mu g/m^3$ . Annually, there were four	
Timeframe: 2000-2001	Number of Sources/Species: 8 sources and 32 species	main sources: secondary nitrate from local and upwind sources of NO <sub>X</sub> and NH <sub>3</sub> (36%), coal combustion including regional transport and secondary sulfates (26%), secondary organic	
Frequency: Daily 24-hour averages	Sources: Coal combustion, secondary nitrate,	aerosols from a variety of precursor sources (15%), and motor vehicle traffic including road dust (10%). Four minor sources	
Data Source: Special study	secondary organic aerosols, motor vehicle traffic, road salt, primary smelters, primary coal and oil	were road salt and three types of primary PM <sub>2.5</sub> associated with smelters, coal and oil combustion, industry and local construction. Overall, about 40% of the total PM <sub>2.5</sub> was	
Data Description: Speciated PM <sub>2.5</sub> data, including	combustion, primary industry, and local construction.	estimated to be from motor vehicle-related emissions (including road salt and nitrates).	
Speciated PM <sub>2.5</sub> data, including 15 trace metals, 7 inorganic ions, 5 water-soluble organic acids, and elemental and organic carbon (separated into 4 temperature fractions).  Analytical techniques included: gravimetric, inductively coupled plasma (ICP)-atomic emission spectroscopy, ICP-mass spectroscopy, ion chromatography, capillary electrophoresis, and thermal optical transmission.	Methodology: Water-soluble, low molecular weight organic acids were included in PMF analysis, improving its ability to identify sources and avoid overestimating secondary coal sources. Several statistical tests—factor indication function, F-test statistics, and cluster analysis with Pearson coefficient—were used to select the optimal number of sources. PMF was run separately for the full dataset, for the summer (May-Oct), and for the winter (Nov-Apr).	Temporal Signals:  In the summer, the secondary coal source is 2 times larger than in the winter, due to photochemical activity and meteorological conditions (transport). Annually, organics contribute ~35% to the total PM <sub>2.5</sub> . Secondary organic aerosols were significantly higher in the summer (23%) compared to the winter (6%), likely from incomplete combustion, biogenic emissions, and biomass burning enhanced by photochemistry and meteorological conditions. The secondary ammonium nitrate source is twice as large in the winter (50%) than the summer (21%), creating a wintertime haze. In the winter, the road salt source also increases.	
	Other Analyses: Back trajectories were run on 6 days	Transboundary Transport: High sulfate days were generally from sources to the south,	

with highest concentrations of several

sources.

<b>Study Reference:</b> Lee, P.K.H., J.R. Brook, E. Dabek-Zlotorzynska, and S.A. Mabury (2003b). Identification of the major sources contributing to PM <sub>2.5</sub> observed in Toronto. Submitted to <i>Environmental Science &amp; Technology</i> .			
<b>Type of Study:</b> Peer reviewed re	esearch	Funding Source: Toxic Substances Research Initiative,	
		Government of Canada	
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations	
		Water-soluble, low molecular weight organic acids should be included in PMF to improve apportionment of secondary sulfates. More research is needed to determine the processes creating the organic acid source and how it related to secondary coal sources.	
		Upcoming Studies/Planned Work: UNMIX analysis has been done for these data and will be reported in the near future. PMF on 2001-2002 data just beginning. In the longer term, the PMF results for 2000-2003 will be linked with acute health effects studies.	

Study Reference: Lewis, C.W., G.A. Norris, R.C Henry, and T.L. Conner (2002). Source Apportionment of Phoenix PM2.5 Aerosol with the UNMIX Receptor Model. Journal of the Air and Waste Management Association (paper accepted September 2002, in press)

**Type of Study:** Peer reviewed research **Funding Source:** U.S. EPA Office of Research and

Type of Study: Peer reviewed research		Development	
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations	
Site Location(s) and Type(s): Phoenix, AZ  Timeframe: 1995-1998  Frequency: Daily 24-hour integrated PM samples  Data Source: Special study  Data Description: PM <sub>2.5</sub> mass, speciated PM <sub>2.5</sub> , OC, EC, and K <sub>W</sub> (parameter representing K from wood burning) analyzed with gravimetric, XRF spectroscopy,	Source Apportionment Tool (s): UNMIX  Number of Sources/Species: 5 sources and over 10 species  Sources: Gasoline engines, diesel engines, secondary sulfate, crustal/soil, and vegetative burning.  Methodology: Data placed in base data set and subject to screening and outlier evaluation, including identifying a potential data artifact with the organic carbon samples. UNMIX was used to determine contributing sources using PM a mass elemental species (Si. S. K.)		
and thermal optical transmission. EPA AIRS data were used for criteria pollutant data (CO, NO <sub>2</sub> , and SO <sub>2</sub> ).	Ca, Mn, Fe, Al, Br), K <sub>w</sub> , OC, EC, and gaseous species. Gasoline and diesel engine split was accomplished by the relative abundances of OC and EC, as well as correlation with CO and NO <sub>2</sub> , the presence of Mn, and the weekdayweekend variations.  Other Analyses:  Scanning electron microscopy (SEM) of a limited number of filter samples to identify low-strength sources	identifying the Mn as likely from iron foundries instead of mobile diesel emissions.  The results of this study were found to be generally consistent to an earlier analysis of the same data using PMF (Ramadan, et al., 2000). Average PM <sub>2.5</sub> concentration differed by less than 4%. The best agreement was for secondary sulfate and vegetative burning. Diesel sources did not correlate well between the UNMIX and PMF results and gasoline results were mixed; this may be due to Mn sources. PMF identified more sources than UNMIX, including sea salt and nonferrous metal smelting.  Temporal Signals:	

Study Reference: Lewis, C.W., G.A. Norris, R.C Henry, and T.L. Conner (2002). Source Apportionment of Phoenix PM2.5 Aerosol with the UNMIX Receptor Model. Journal of the Air and Waste Management Association (paper accepted September 2002, in press) Type of Study: Peer reviewed research Funding Source: U.S. EPA Office of Research and Development **Site and Data Specifications Source Apportionment Method Findings and Recommendations** In the winter, gasoline engines dominate (42%). In the summer, secondary sulfate (29%) and crustal/soil (26%) dominate (gasoline engines are 20% in the summer). Transboundary Transport: None mentioned. Recommendations: UNMIX and PMF did not agree in preliminary independent evaluation. Thus, they recommend a concurrent iterative application of UNMIX and PMF. Due to the increasing power and complexity of these models, they should be used with caution, experience, and cooperation. Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Liu, W., P.K. Hopke, Y. Han, S.-M. Yi, T.M. Holsen, S. Cybart, K. Kozlowski, and M. Milligan (2003a). "Application of receptor modeling to atmospheric constituents at Potsdam and Stockton, NY." Submitted for publication.

**Type of Study:** Peer review research

Funding Source: New York State Energy Research and Development Authority

Development Authority		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Stockton, NY (rural) Potsdam, NY (rural)	Source Apportionment Tool (s): PMF Number of Sources/Species:	Results:  No total mass concentration values were provided; only percent contribution from each source.
Timeframe: May 15-August 31, 2000 and May 15-August 31, 2001	Stockton: 6 sources and 44 species Potsdam: 7 sources and 44 species Sources:	For Stockton, the source contributions were: secondary sulfate (56%), secondary nitrate (2%), soil (24%), wood smoke (2%), zinc smelter (4%), and copper smelter (12%).
Frequency: Daily 23-hour integrated samples	Stockton: Secondary sulfate, secondary nitrate, soil, wood smoke, zinc smelter, and copper smelter.	For Potsdam, the source contributions were: secondary sulfate (56%), secondary nitrate (9%), soil (15%), wood smoke (9%), zinc smelter (2%), copper smelter (2%), and nickel smelter
Data Source: Special study Data Description:	Potsdam: Secondary sulfate, secondary nitrate, soil, wood smoke, zinc smelter, copper smelter, and nickel smelter.	(4%).  The secondary sulfate, nitrate, and soil sources contained very little of the PAH species. The wood smoke and smelter
Speciated PM <sub>2.5</sub> , vapor phase mercury, and polycyclic aromatic hydrocarbon (PAH) data. Analysis of PM <sub>2.5</sub> data	Methodology: PMF was used to determine sources. Missing data was replaced by geometric mean of corresponding elements. Half	sources had many PAH species associated with them.  Temporal Signals: Sulfate levels differed between the two summers and between the two cities, primarily due to meteorological differences.
was conducted with ion chromatography. Mercury was analyzed with thermal desorption and cold vapor	the detection limit was used for data below detection. PMF was operated in the robust mode.	Transboundary Transport: Sulfate for the two locations had a large common source area from the lower Great Lakes to south of the Ohio River Valley.
desorption and cold vapor atomic fluorescence spectrometry. PAHs were analyzed using gas chromatography/mass spectrometry.	PMF was used with 43 input species not including total mass: Al, Si, P, S, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Se Br, Sr, Pb, black carbon, NO <sub>3</sub> , Hg, and 21 PAHs. Total mass was apportioned by regression.	Nitrate also appeared to have a common source in the Midwest and in southwestern New York. Source regions for soil included the eastern part of the agricultural Midwest and eastern states from Pennsylvania to Virginia. The wood smoke source was from the Great Lake area. Smelter sources were from various regions in Canada.
	Other Analyses: HYSPLIT and potential source	Recommendations: None mentioned.

Study Reference: Liu, W., P.K. Hopke, Y. Han, SM. Yi, T.M. Holsen, S. Cybart, K. Kozlowski, and M. Milligan (2003a). "Application of receptor modeling to atmospheric constituents at Potsdam and Stockton, NY." Submitted for publication.  Type of Study: Peer review research  Funding Source: New York State Energy Research and		
		Development Authority
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	contribution function (PSCF) were used	Upcoming Studies/Planned Work:
	to determine probable location.	None mentioned.

**Study Reference:** Liu, W., P.K. Hopke, and R.A. VanCuren (2003b). "Origins of Fine Aerosol Mass in the Western United States Using Positive Matrix Factorization." Submitted to *Journal of Geophysical Research*.

<b>Type of Study:</b> Peer reviewed rese		Funding Source: California Air Resources Board
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): Crater Lake National Park, OR; Lassen Volcanic National Park, CA (both elevated remote sites)  Timeframe: 1992-2000  Frequency: Integrated 24-hour samples every Wednesday and Saturday  Data Source: IMPROVE network  Data Description: Speciated PM <sub>2.5</sub> data. Analytical techniques included gravimetric, X-ray fluorescence, proton elastic scattering analysis, proton induced X-ray emission, ion chromatography, and thermal-optical reflectance.	Source Apportionment Tool (s):     PMF  Number of Sources/Species:     Crater Lake NP: 7 sources and 20 species     Lassen Volcanic NP: 6 sources and 20 species  Sources:     Crater Lake NP: Wind-blown Asian dust, secondary sulfate, wood burning, nitrate, sea salt, mixed motor vehicle, and a Zn-Cu source.  Lassen Volcanic NP: Wind-blown Asian dust, secondary sulfate, wood burning, nitrate, sea salt, and mixed motor vehicle.  Methodology:     PMF was used to determine sources.     Missing data was replaced by geometric mean of corresponding elements. Half the detection limit was used for data below detection. PMF was operated in the robust mode.  PMF was used with 20 input species not including total mass: Al, As, Br, Ca, Cr, Cu, Fe, K, Mn, Ni, P, Pb, Rb, S, Si, Sr, Ti, V, Zn, and Zr. Total mass was apportioned by regression.	Results:  The goal of the study was to evaluate the impact of Asian dust on air quality. More impact from Asian dust was found than had previously been reported. No total mass concentration values were provided; only percent contribution from each source. Also, percent contributions in the figures did not match values reported in the text. Values from the figures are reported here.  For Crater Lake NP, the source contributions were: wind-blown Asian dust (12%), secondary sulfate (15%), wood burning (46%), nitrate (2%), sea salt (4%), mixed motor vehicle (12%), and a Zn-Cu source (3%).  For Lassen Volcanic NP, the source contributions were: wind-blown Asian dust (11%), secondary sulfate (26%), wood burning (48%), nitrate (6%), sea salt (1%), and mixed motor vehicle (1%).  The chemical composition of the Asian dust compared fairly well with its known composition except for S, Ca, and Si. Sulfur was 6-7 times higher than reference materials and was likely from accumulation during transport through industrial areas in eastern China. Ca and Si were less than reference materials likely due to deposition.  The sulfate source may be from both industrial and marine sources. The wood burning source may be from local residential wood burning and forest fires.  Temporal Signals:  Wind-blown Asian dust occurs most frequently in spring between March and May, including some high peaks during

dust events. The motor vehicle source is higher in the summer

Study Reference: Liu, W., P.K. Hopke, and R.A. VanCuren (2003b). "Origins of Fine Aerosol Mass in the Western United States Using Posit Matrix Factorization." Submitted to <i>Journal of Geophysical Research</i> .  Type of Study: Peer reviewed research  Funding Source: California Air Resources Board		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	Other Analyses: HYSPLIT was mentioned but not used extensively.	during tourist season. The Zn-Cu source occurred in sporadic events.  Transboundary Transport: The wind-blown Asian dust source is from trans-Pacific transport.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Long, R.W. (2002). Measurement of PM<sub>2.5</sub>, including semi-volatile components, in the EPA EMPACT and STAR programs: Results from the Salt Lake City, Bountiful, and Lindon, Utah, studies and implications for public awareness, health effects, and control strategies [dissertation]. Provo (UT): Brigham Young University.

[dissertation]. Provo (UT): Brigham Young University. <b>Type of Study:</b> Ph.D. dissertation <b>Funding Source:</b> None mentioned		
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
	Source Apportionment Method  Source Apportionment Tool: UNMIX  Number of Sources/Species: 3-4 sources per site per campaign.  Sources: Secondary wood smoke, primary wood smoke, primary oil refinery, primary mobile, primary automotive, primary diesel.  Methodology: The one hour average data were analyzed using UNMIX for each of the sampling campaigns. During several sampling periods at the Hawthorne site, UNMIX distinguished gasoline and diesel emission using semi-volatile species as well as EC, CO, and NO <sub>X</sub> concentrations.  Other Analyses: None.	
STAR site (Bountiful)  Data Description:  Total PM <sub>2.5</sub> , non-volatile PM <sub>2.5</sub> , semi-volatile organic matter, elemental carbon, UV, and gaseous data from a range of real-time ambient mass sampling instruments. A PC-BOSS sampler was used to determine particle composition.		high during the Olympic games.  high during the Olympic games.  Mobile sources showed a diurnal pattern with morning and evening.  Transboundary Transport: None mentioned.  Recommendations: Further study of real-time monitors should be
		Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Lowenthal, D.H. and K.A. Rahn (1989). The relationship between secondary sulfate and primary regional signatures in northeastern aerosol and precipitation. *Atmospheric Environment*, **23**, 1511-1515.

Type of Study: Peer reviewed research

**Funding Source:** U.S. EPA, U.S. Department of Energy, and Ohio Electric Utility Institute

Findings and Recommendations
Results:  Sulfate levels were determined to be linearly related to the signatures. Local aerosol levels are no more variable that distant sources. Sulfates were confirmed to have regional signatures on a scale of 300-500 km in size and not "superregional" (e.g., they are not well-mixed on a larger scale). In the summer, 56% of the sulfate was from the northeast and 44% from the midwest. In the winter, 61% of the sulfate was from the northeast, 33% from the midwest, and 6% from smelters.  Temporal Signals:  Sulfates in both aerosols and precipitation are generally higher in the summer due to higher oxidation rates, except for northeastern effective sulfate in rain, which is higher in winter than summer due to higher local emissions.  Transboundary Transport:  Significant transport from the midwest and some transport from smelters in Canada.  Recommendations:  Source apportionment can be accomplished with sulfate aerosols.  Upcoming Studies/Planned Work:  None mentioned.
T R

**Study Reference:** Maykut, N.N., J. Lewtas, E. Kim, and T.V. Larson (2003). Source apportionment of PM<sub>2.5</sub> at an urban IMPROVE site in

Seattle, WA. Submitted to *Environmental Science & Technology*.

**Type of Study:** Peer reviewed research

Funding Source: U.S. EPA and University of Washington/EPA
Northwest Research Center for Particulate Air Pollution and

Health

#### **Site and Data Specifications**

# Site Location(s) and Type(s): Seattle, WA (urban)

# Timeframe: 1996-1999

# Frequency:

24 hour averages twice weekly (289 samples total)

#### Data Source:

**IMPROVE** site

#### Data Description:

Total PM<sub>2.5</sub>, speciated PM<sub>2.5</sub> data collected with the IMPROVE protocol, including 4 temperature resolved particulate organic carbon fractions and 3 elemental carbon fractions. Analysis included proton elastic scattering, particle induced X-ray emission, X-ray fluorescence, and thermal/optical reflectance.

#### **Source Apportionment Method**

Source Apportionment Tool: PMF, UNMIX, and CMB

# Number of Sources/Species:

PMF: 8 sources and 32 species UNMIX: 6 sources and 15 species CMB: 7 sources and 23 species

#### Sources:

PMF: gasoline, diesel, vegetative, fuel oil, soil, marine, Na-rich, and secondary UNMIX: fuel oil, vegetative, gasoline, diesel, soil, and marine.

CMB: vegetative burning, mobile, soil, Na-rich, fuel oil, marine, and sulfate.

# Methodology:

PMF, UNMIX, and CMB were used on the same data in order to compare results. Total annual contributions were calculated as well as summer (Mar-Sept), winter (Oct-Feb), and day of week (Wednesday and Saturday).

PMF: Due to data below detection limits, Mg, P, Zr, and organic pyrolitic carbon were not used in the model. Chlorine (instead of chlorine ion) and sulfate ion (instead of elemental sulfur) were used. Trace elements for various combustion sources were used to

# Findings and Recommendations

#### Results:

The mean total  $PM_{2.5}$  mass is  $8.9 \,\mu g/m^3$ . While the results between the models were similar, the overall results varied considerably between them. The models responded differently to certain elements such as Mn, As, and carbon fractions. The top sources were:

vegetative burning (CMB 16% UNMIX 37% PMF 28% inventory 28%)

mobile sources (CMB 44% UNMIX 28% [diesel 19% gas 9%] PMF 22% [diesel 18% gas 4%] inventory 23% [diesel 18% gas 5%])

secondary sulfate (CMB 17% UNMIX NA PMF 18% inventory NA)

fuel oil (CMB NA UNMIX 15% PMF 10% inventory 2%) soil (CMB 4% UNMIX 6% PMF 14% inventory 3%) marine/sea salt (CMB 7% UNMIX 12% PMF 3% inventory NA)

Others that were smaller sources and were not consistent found by all models included industry (CMB 7% inventory 3%), Na rich (PMF 5%), secondary nitrate (CMB 4%), and combined sulfate, nitrate, and marine (inventory 34%).

When compared to the 1996 King County emissions inventory, PMF and UNMIX values were in better agreement than CMB. The exception was smaller sources from specific industries that PMF and UNMIX could not resolve, but CMB could point directly toward once the profile was loaded. The major difference between CMB and PMF/UNMIX models was the

Study Reference: Maykut, N.N., J. Lewtas, E. Kim, and T.V. Larson (2003). Source apportionment of PM<sub>2.5</sub> at an urban IMPROVE site in Seattle, WA. Submitted to *Environmental Science & Technology*. Type of Study: Peer reviewed research Funding Source: U.S. EPA and University of Washington/EPA Northwest Research Center for Particulate Air Pollution and Health **Site and Data Specifications Source Apportionment Method Findings and Recommendations** resolve diesel from other mobile relative contribution of vegetative burning versus motor vehicles, which may be due to the lack of local source profiles sources. for the CMB model and the inability to use temperature UNMIX: Certain species for certain resolved carbon fractions. days were removed to improve results, specifically K for fireworks, Al, Si, S, Temporal Signals: Vegetative burning was significantly higher and diesel and Fe for an Asian dust event, and lead emission slightly higher in the winter. Soil, fuel oil, and and organic carbon on two days for unexplained reasons. Temperature secondary sulfates were slightly higher in the summer. A resolved organic and elemental carbon comparison of results between Wednesdays and Saturdays showed that diesel is significantly higher on Wednesdays over fractions were used to enable UNMIX to separate diesel from other mobile Saturdays and vegetative burning is higher on Saturdays than Wednesdays. sources. CMB: Sources were based on a 1996 Transboundary Transport: inventory of PM<sub>2.5</sub> emission in King Major transboundary event (Asian dust) on April 29, 1998 was County. Source profiles came from removed from analysis. No other transboundary transport other published work, including a discussed. composite gasoline/diesel profile from Recommendations: Phoenix. Carbon fractions were not None mentioned. used in the CMB analysis due to insufficient information. Upcoming Studies/Planned Work: None mentioned. Other Analyses: Results were compared with the 1996

King County emissions inventory.

Study Reference: Poirot, R.L., P.R. Wishinski, P.K. Hopke, and A.V. Polissar (2001). Comparative application of multiple receptor methods to identify aerosol sources in northern Vermont. Environmental Science & Technology, 35, 4622-4636.

Funding Source: None mentioned

inputs also included Zn, Pb, Br, Cu, Ti,

Na, Al, V, Sr, Cr, Rb, Mg, Zr, Mo, Cl,

Two backward trajectory techniques:

potential source contribution function

Type of Study: Peer reviewed research		
Site and Data Specifications	Source Apportionment Method	
Site Location(s) and Type(s): Underhill, VT (rural)	Source Apportionment Tool (s): UNMIX and PMF	
Timeframe: 1988-1995 Frequency:	Number of Sources/Species: UNMIX: 7 sources and 11 species PMF: 11 sources and 27 species	
Integrated 24-hour samples every Wednesday and Saturday, plus every sixth day.	Sources: 7 sources (PMF and UNMIX): midwest summer coal, midwest winter coal,	
Data Source: IMPROVE network and the NESCAUM Regional Particle Monitoring Network	wood smoke, east coast oil, soil, Canadian Mn, and Canadian smelter. Plus 4 additional sources (PMF only): Zn-Pb, Na-S, Cu, and salt.  Methodology: The two source apportionment mathematical models and two back trajectory methods were run on the sai dataset in order to determine usefulne and limitations of these methods. Tot mass was included as UNMIX input. PMF mass attribution was by regression. Both PMF and UNMIX h inputs of S, Fe, K, H, Ca, Si, Se, Ni, Mn, As, and light absorption. PMF	
Data Description:  Speciated PM <sub>2.5</sub> data using samplers and analysis similar to those in the IMPROVE network. Analytical techniques included gravimetric, laser integrating plate, proton elastic scattering analysis, and proton induced X-ray emission/X-ray fluorescence.		

and P.

Other Analyses:

# **Findings and Recommendations**

#### Results:

The mean reconstructed mass for PMF was 7.9 µg/m<sup>3</sup> and 8.4 µg/m<sup>3</sup> for UNMIX. Both models (PMF and UNMIX) reproduced the daily mass results reasonably well. The average mass of the source contributions were midwest summer coal (PMF 53% UNMIX 55%), wood smoke (PMF 15% UNMIX 16%), midwest winter coal (PMF 8% UNMIX 14%), east Coast oil (PMF 7% UNMIX 8%), Canadian Mn sources (PMF 2% UNMIX 4%), soil (PMF 4% UNMIX 2%), and Canadian smelter (PMF 1% UNMIX 1%). PMF also identified sources of Zn-Pb (7%), Cu (2%), Na-S (<1%), and salt (<1%). Midwest summer coal accounts for about half the average mass.

The trajectory analysis provided input into the locations of these sources. For example, using probability fields, they were able to identify which smelter had the most influence, that its impact was very episodic, and could see the reductions in emissions from improvements in their emissions control technologies in 1990. They also reviewed probability fields for each source for the 10% of days with the highest concentration, as confirmation of the source location and type.

Convergent results between the two models were the identification of 7 similar sources with similar trajectories. Several divergent results were avoided by adjusting the modeling approach and treatment of the raw data. PMF identified 4 additional sources over UNMIX and apportioned the total mass over those 11 sources, resulting in different mass results for the 7 common sources. The models handled unique events (in this case, a nearby fire and subsequent construction in spring 1991) in different manners and with divergent results. **Study Reference:** Poirot, R.L., P.R. Wishinski, P.K. Hopke, and A.V. Polissar (2001). Comparative application of multiple receptor methods to identify aerosol sources in northern Vermont. *Environmental Science & Technology*, 35, 4622-4636.

Type of Study: Peer reviewed research

Funding Source: None mentioned

Type of Study: Peer reviewed res	search	Funding Source: None mentioned
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
	(PSCF) and residence-time analysis (RTA).	Trace metals (e.g., Al) also produced problems with either rejection of the data as input (UNMIX) or misquantified results (PMF), although this appears to be a date artifact related to the Al detection limit. Both models had some difficulty in identifying secondary sulfate although were relatively robust in the midwest coal sources.
		Temporal Signals: Two coal sources (identified by their different S:Se ratios) had summer and winter maximums, respectively. East coast oil and wood smoke both had winter peaks and the soil source peaked in the spring. The other sources did not have strong seasonal patterns.
		Transboundary Transport:  Due to its location near the Canadian border, two sources were from cross-border transport (the Mn and smelter sources).  Two of the largest sources (summer and winter coal) were transported from the midwest, presumably from power plants.
		Recommendations:  Both models and both trajectory analyses should be used for inter-comparison of results and should not be considered stand-alone techniques at this time. Both techniques are sensitive to systematic errors and biases in the input data, including decisions made in the treatment of missing data and data below detection limits.
		Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Polissar, A.V., P.K. Hopke, and R.L. Poirot (2001). Atmospheric aerosol over Vermont: Chemical composition and sources. *Environmental Science & Technology*, **35**, 4604-4621.

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site And Data Specifications  Site Location(s) and Type(s): Underhill, VT (rural)  Timeframe: 1988-1995  Frequency: Integrated 24-hour samples every Wednesday and Saturday, plus every sixth day.  Data Source: IMPROVE network and the NESCAUM Regional Particle Monitoring Network  Data Description: Speciated PM <sub>2.5</sub> data. Analytical techniques included gravimetric, laser integrating plate, proton elastic scattering analysis, and proton induced X-ray emission/X-ray fluorescence.	Source Apportionment Tool (s):     PMF  Number of Sources/Species:     11 sources and 27 species  Sources:     Salt, Na-S, Canadian smelting, Cu smelting, soil, Canadian Mn, Zn-Pb, midwest summer coal, east coast oil, midwest winter coal, and wood smoke.  Methodology:     PMF analysis was performed to determine sources using 27 sources (Al, As, BC, Br, Ca, Cl, Cr, Cu, Fe, H, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Si, Sr, Ti, V, Zn, and Zr). PSCF and back trajectory analysis was then conducted to determine direction and probability information about the sources. Total mass was excluded from the models.  Other Analyses:     Potential source contribution function (PSCF) analysis to identify possible source areas.  CAPITA Monte Carlo trajectory model	Results:  Total average fine aerosol mass was 6.4 μg/m³ and the concentration ranged from 0.2-51.1 μg/m³. Percentage of each source for the whole period were: midwest summer coal (51%), wood smoke (15%), midwest winter coal (7%), Zn-Pb (7%), east coast oil (7%), soil (4%), Canadian Mn (2%), Cu smelting, Canadian smelting, Na-S, and salt (all less than 2%), and undetermined (4%).  The report states that 87% of fine mass concentration was from 4 sources: fuel consumption, local wood smoke, municipal waste incineration, and secondary sulfate production. [Note: The paper does not clearly state how these sources directly relate to the PMF results, but it is likely the five largest PMF sources listed above.]  Black carbon was from residential wood combustion in northern New England and southwestern Quebec. The coal combustion and Zn-Pb sources were from the midwestern U.S. Pb-Mn was also from the midwest as well as Montreal. Oil combustion was from the east coast of the U.S. The As source was Canadian nickel smelters with some additional contribution from power plants south and west of the site. Windblown dust was primarily from areas to the north.  Temporal Signals:  Sulfur and total fine particle mass had maxima in the summer and minima in the winter. Black carbon had no seasonal pattern. Most anthropogenic sources had maxima in the winter

Study Reference: Polissar, A.V., P.K. Hopke, and R.L. Poirot (2001). Atmospheric aerosol over Vermont: Chemical composition and sources.  Environmental Science & Technology, 35, 4604-4621.  Type of Study: Peer reviewed research  Funding Source: National Science Foundation		
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
		Recommendations: Combined use of PMF and PSCF was effective in identifying aerosol emissions and their sources.  Upcoming Studies/Planned Work: None mentioned.

Study Reference: Rahn, K.A. and D.H. Lowenthal (1984). Elemental Tracers of Distant Regional Pollution Aerosols. <i>Science</i> , 223, 132-139  Type of Study: Peer reviewed research  Funding Source: None mentioned		
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): Narragansett, RI (rural) Underhill, VT (rural) Barrow, AK (rural) (+3 in Europe) Timeframe:	Source Apportionment Tool (s): Regional elemental tracer analysis Number of Sources/Species: 7 tracer elements and 6 U.S. regional signatures	Results: At the Underhill site, the signatures indicated 40% of the PM was from New England, 30% from other east coast cities, and 25% from the interior. At Narragansett, about 75% was from the coastal cities and about 25% from the interior. Sulfate signatures for the Underhill site were estimated to be
Samples were taken between 1976 and 1982 depending on the site.  Frequency: Signatures were developed by	Sources: Tracer elements: As, Sb, Se, Zn, In, noncrustal V, and noncrustal Mn Regional source areas: New England, Boston, New York City, Washington	21 µg/m <sup>3</sup> for the interior, 7 µg/m <sup>3</sup> for the mid-Atlantic/ Washington region, and 3 µg/m <sup>3</sup> for the local New England region, which was estimated to be accurate to about 25%. A similar approach at the Narragansett site was not as accurate, likely due to closer sources.
signatures were developed by taking between 31 and 79 samples depending on the site. Sampling was done in campaigns of various lengths and seasons depending on the site.  Data Source: Special study  Data Description: Filter samples of total aerosol analyzed for tracer elements, As, Sb, Se, Zn, In, noncrustal V, and noncrustal Mn. No further	DC, interior, Canadian smelter/southern Ontario, and 6 European source areas.  Methodology: By assessing the proportions of elements in aerosols, some apportionment can be made of the source area of the pollutants. Regional aerosol signatures are mixtures of point sources and are determined empirically by taking modal information from several sites in the region. The modes of the ratios of 6 tracer elements to Se are used to determine a regional	Differences in elemental tracers are possible to measure on a regional scale. The best regional signatures are made up of a few elements with the greatest tracer power. Requirements for tracers are: pollution-derived, able to be sampled and measured accurately, stable homogeneous emission, and present on particles that can be transported long distances.  The study found that As/Se and noncrustal V/Se were the ratios with the greatest discriminatory power; Zn/Se had somewhat less power, and the other three ratios contributed little. They tested other elements for their potential as tracers with moderate results and some questions remaining about their ability to be used with fine particles versus coarse particles.
information on analytical technique was provided.	signature for the secondary species sulfate was also developed through regression analysis with the 7 tracers.  Other Analyses:	Temporal Signals:  The mix of sources can vary significantly due to large scale meteorology, creating seasonal changes and pollution episodes.
	None.	Transboundary Transport:  Many of the Barrow and other Arctic signatures were not from

<b>Study Reference:</b> Rahn, K.A. and D.H. Lowenthal (1984). Elemental Tracers <b>Type of Study:</b> Peer reviewed research		cers of Distant Regional Pollution Aerosols. <i>Science</i> , <b>223</b> , 132-139 <b>Funding Source:</b> None mentioned
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
		North American sources, which indicates that pollution aerosols in the Arctic are strongly influenced by Eurasian transport.
		Recommendations: While some elements were found to have good potential as tracers, they recommend measuring as many elements as possible (both natural and pollution derived) in order to use them to monitor more transient or unique events such as dust storms, volcanic eruptions, or non-routine point source emissions.
		Upcoming Studies/Planned Work:  More work finding better tracer elements, improving statistical validity, evaluating better size-segregated sampling, and refining tracing secondary species.

**Study Reference:** Ramadan, Z., B. Eickhout, X.-H. Song, L.M.C. Buydens, and P.K. Hopke (2003). "Comparison of Positive Matrix Factorization and Multilinear Engine for the source apportionment of particulate pollutants." *Chemometrics and Intelligent Laboratory Systems*, 66, 15-28.

<b>Type of Study:</b> Peer reviewed research	Funding Source: U.S. EPA
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Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Phoenix, AZ Timeframe:	Source Apportionment Tool (s):  ME  Number of Sources/Species:	Results: With the DFPSS / ME analysis, the major fine PM sources were motor vehicles and coal fired power plants. Next highest
DFPSS: 1995-1998 DICHOT: 1996-1998	DFPSS / ME: 7 sources DFPSS & DICHOT / ME: 8 sources	were soil and biomass burning. No total masses or percents of sources were indicated.
Frequency: DFPSS: Daily integrated 24-hour samples DICHOT: Every third day 24-hour integrated sample	Sources: DFPSS / ME: Heavy duty diesel, biomass burning, soil, non-ferrous smelting, coal power plant, aged sea salt, and motor vehicles.	The three-way analysis was not as successful as hoped. Soil, motor vehicle, coal power plant, and biomass burning profiles were recognized. An incinerator profile was identified. However, there were inconsistencies with the smelter and other sources that could not be explained.
Data Source: Special study  Data Description: Speciated PM <sub>2.5</sub> data from both a dual fine particle sequential sampler (DFPSS) and a dichotomous sampler (DICHOT). Analytical techniques included dispersive X-ray spectrometry and thermal optical transmission technique.	DFPSS & DICHOT / ME: Soil, biomass burning, non-ferrous smelting (DICHOT), non-ferrous smelting (DFPSS), incinerator, coal power plant, motor vehicles, and soil.  Methodology: PMF and ME were first used with the DFPSS data. The PMF analysis was presented in Ramadan et al. (2000).  A three way analysis was conducted with the DFPSS and DICHOT data to try and confirm the results with the DFPSS data. In order to complete the DICHOT dataset, Na, OC, and EC from the DFPSS were used.	Temporal Signals:    Motor vehicle and biomass burning sources peaked in the winter and coal fired power plant sources peaked in the summer.  Transboundary Transport:    None mentioned.  Recommendations:    The three way analysis was not successful.  Upcoming Studies/Planned Work:    None mentioned.
	Other Analyses: None mentioned.	

**Study Reference:** Ramadan, Z., X.H. Song, and P.K. Hopke (2000). Identification of sources of Phoenix aerosol by positive matrix factorization. *Journal of the Air and Waste Management Association*, **50**, 1308-1320.

Type of Study: Peer reviewed rese	earch	Funding Source: U.S. EPA
<b>Site and Data Specifications</b>	Source Apportionment Method	Findings and Recommendations
Site and Data Specifications  Site Location(s) and Type(s): Phoenix, AZ  Timeframe: 1995-1998  Frequency: Fine PM: Daily integrated 24-hour samples  Coarse PM and second set of fine PM: Every-third-day 24-hour integrate samples  Data Source: Special study		Findings and Recommendations  Results:     Major fine PM sources were motor vehicles, vegetation burning, and coal-fired power plants. Major sources of coarse PM are soil and construction. No total masses or percent for sources was indicated.  Temporal Signals:     The major PM sources peaked in the winter, which corresponds to peak tourist season and fireplace use.  Transboundary Transport:     None mentioned.  Recommendations:     None mentioned.  Upcoming Studies/Planned Work:
Data Description: Two sets of fine particle samples from different samplers (DFPSS and DICHOT) and one set of coarse PM. Data were speciated for 46 chemical species with X-ray fluorescence and thermal optical transmission.	Methodology: PMF was applied to all three datasets and the results were compared. PMF used 46 chemical species not including total mass: Na Mg Al Si P S Cl K	None mentioned.

Study Reference: Song, X.H., A.V. Polissar, and P.K. Hopke (2001). Sources of fine particle composition in the northeastern US. Atmospheric

Environment, 35, 5277-5286.

Type of Study: Peer reviewed research

**Funding Source:** California Air Resources Board, New Jersey Department of Environmental Protection, and U.S. EPA.

# **Site and Data Specifications**Site Location(s) and Type(s):

Washington, DC (urban)
Brigantine, NJ (rural)
Underhill, VT (rural)

#### Timeframe:

1988-1999 (Washington) 1992-1999 (Brigantine) 1988-1995 (Underhill)

#### Frequency:

Integrated 24-hour samples every Wednesday and Saturday, plus every sixth day.

#### Data Source:

IMPROVE network (Washington and Brigantine) and the NESCAUM Regional Particle Monitoring Network (Underhill)

#### Data Description:

Speciated PM<sub>2.5</sub> data and analytical techniques including gravimetric, laser integrating plate, proton elastic scattering analysis, and proton induced X-ray emission/X-ray fluorescence.

# Source Apportionment Method

Source Apportionment Tool (s): PMF

## Number of Sources/Species:

8 sources, 30 species for Washington 9 sources, 30 species for Brigantine 11 sources, 27 species for Underhill

#### Sources:

Six of the sources had common chemical composition and seasonality between the sites: secondary sulfate, coal combustion, oil combustion, soil, incinerator, and sea salt. Washington also had nitrate and motor vehicle sources; Brigantine had nitrate, motor vehicle, and wood smoke; and Underhill had wood smoke, Canadian Mn, Canadian Cu smelter, Canadian Ni smelter, and a second salt source.

## Methodology:

PMF was used to analyze the composition data at all three sites. Data below detection limit used half the concentration value. Missing data were replaced with the geometrical mean. Some adjustments to this process were made to the Underhill data.

## Other Analyses:

None.

# **Findings and Recommendations**

#### Results:

The total mass was not provided, but summing the mass values in the results tables indicates average masses of about  $18 \mu g/m^3$  (Washington),  $11 \mu g/m^3$  (Brigantine), and  $8 \mu g/m^3$  (Underhill). Overall, the secondary sulfate source contributes the most mass at each site: Washington (47%), Brigantine (55%), and Underhill (51%). In Washington, nitrates are also a major contributor (almost 20%) and at Underhill wood smoke is 14%.

The secondary sulfate source has a high concentration of S and is related to coal combustion, while the coal combustion source has S and Se. The differences in atmospheric chemistry for these signatures indicate two types of emission sources.

The oil combustion source contains Ni and V and probably represents fuels for utility and industrial sources in the northeast. Soil represents windblown dust. Incinerator sources were attributed to municipal solid waste incineration and non-ferrous metal smelting.

# Temporal Signals:

Secondary sulfate source has high concentrations in the summer and the coal combustion source peaks in the winter. Oil combustion and incineration sources peak in the winter. Soil and sea salt exhibited no seasonal trends.

## Transboundary Transport:

Emissions from Canadian smelters and transportation sources are likely.

#### Recommendations:

PMF is a powerful and useful analytical method to extract

<b>Study Reference:</b> Song, X.H., A.V. Polissar, and P.K. Hopke (2001). Sources of fine particle composition in the northeastern US. <i>Atmospheric Environment</i> , <b>35</b> , 5277-5286.		
Type of Study: Peer reviewed research  Funding Source: California Air Resources Board, New Jersey		
		Department of Environmental Protection, and U.S. EPA.
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
		emissions sources from concentration data.
		Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Wishinski, P.R. and R.L. Poirot (1986). Source/receptor relationships for a number of factors contributing to summertime variation in light extinction in northern Vermont, <u>Transactions APCA Spec. Conf. on Visibility Protection: Research and Policy Aspects</u>, P.S. Bhardwaja, Ed., Jackson Hole, WY, pp 807-822.

Type of Study: Conference Proceedings	Funding Source: None mentioned
Site and Data Specifications Source Apportionment Me	thod Findings and Recommendations
Site Location(s) and Type(s): Underhill, VT (rural)  Timeframe: Summer 1983  Frequency: Daily  Data Source: Special study  Data Description: Relative humidity, sulfate, nonsulfate TSP, and aerosol trace elements.  Source Apportionment Tool Principal component analy  Number of Sources/Species: 5 factors and 15 variables  Sources: Coal combustion, oil combusture content, crustal, and temporal to data as Regression analysis against spatial and temporal charat were examined with reside analysis.  Other Analyses: Residence time analysis	Coal combustion in the lower Great Lakes and Ohio River Valley were the primary cause of visual range reduction (47%). The other sources contributed only slightly to light extinction (less than 6% each, 12% for all 4 total).  Oil combustion was associated with urban areas in the northeast, crustal is ubiquitous, moisture is from the south and east, and the industrial signature is from the northern midwest and some from Canada.  Temporal Signals: None, since study was only for one summer.

**Study Reference:** Zheng, M., G.R. Cass, J.J. Schauer, and E.S. Edgerton (2002). Source apportionment of PM2.5 in the southeastern United States using solvent-extractable organic compounds as tracers. *Environmental Science & Technology*, **36**, 2361-2371

Type of Study: Peer reviewed rese	earch	Funding Source: Electric Power Research Institute
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s): Atlanta, GA (urban) N. Birmingham, AL (urban) Gulfport, MS (urban) Pensacola, FL (urban) Centreville, AL (rural) Oak Grove, MS (rural) Yorkville, GA (rural)	Source Apportionment Tool (s):     CMB  Number of Sources/Species:     7 sources and 107 particle phase organic species  Sources:     OC apportionment: Diesel exhaust,	Results:  PM <sub>2.5</sub> mass ranged from 7.9 to 29.2 μg/m³, depending on month and location. The maximum was Atlanta in August and the minimum was Centreville in January. The contributions to PM <sub>2.5</sub> mass concentrations were secondary sulfate, wood combustion, diesel exhaust, secondary ammonium, secondary nitrate, meat cooking, gasoline-powered motor vehicle exhaust, and road dust (exact percentages were not provided).
NW of Pensacola, FL (rural)  Timeframe: April, July, October 1999 and January 2000  Frequency: Daily 24 hour integrated samples in April, July, and October, and every third day in January.	gasoline exhaust, vegetative detritus, meat cooking, road dust, wood combustion, and natural gas combustion (plus unidentified organic carbon source)  Mass apportionment found the same 7 sources, plus other organic matter, secondary sulfate, secondary nitrate, secondary ammonium, and other.  Methodology:	Major sources of final particle organic carbon at the 8 SEARCH sites are (in %OC): wood combustion (25-66%), diesel exhaust (14-30%), meat cooking (5-12%), and gasoline-powered motor vehicle exhaust (0-10%). Small amounts come from natural gas combustion (0-5%), road dust (0.7-2.2%), and vegetative detritus (1.0-2.2%). Highest annual average measured in N. Birmingham followed by Atlanta. Annual averages in urban sites were 1.5 times the rural sites. Urban sites had higher concentrations of diesel and gasoline powered vehicle exhaust.
Data Source: Southeastern Aerosol Research and Characterization (SEARCH) air monitoring network.  Data Description: Speciated PM <sub>2.5</sub> data, including	Used CMB with particle-phase organic compounds as tracers. Source profiles came from existing studies, with two exceptions. The wood combustion profile was developed using woods indicative of the southeast U.S. The road dust profile was a mix of Al-Si	Temporal Signals: Higher contribution from wood combustion in October and January and higher percentage of fine organic carbon in July, possibly due to increase in secondary organic aerosols formation during the summer. Highest concentration from diesel exhaust was in October at 6 of 8 sites and the total gas and diesel contribution increased to 44% in October. Wood
107 particle phase organic species, analyzed with GC/MS. Tracers identified and quantified by gas chromatography/mass	data from Alabama road dust samples with organic data from previous tests in California. Gasoline and diesel were differentiated by OC and EC concentrations and the use of gasoline	smoke dominated (>56%) in January.  Transboundary Transport: None mentioned.  Recommendations:

Study Reference: Zheng, M., G.R. Cass, J.J. Schauer, and E.S. Edgerton (2002). Source apportionment of PM2.5 in the southeastern United States using solvent-extractable organic compounds as tracers. <i>Environmental Science &amp; Technology</i> , <b>36</b> , 2361-2371		
		Funding Source: Electric Power Research Institute
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
spectrometry.	and diesel profiles.	None mentioned.
	Other Analyses:	Upcoming Studies/Planned Work:
None. None mentioned.		

Table A-3. Summary of source apportionment studies involving PM<sub>2.5</sub>: Presentations

Study Reference: Poirot, R.L. and P.R. Wishinski (2001). Application of Combined Mathematical and Meteorological Receptor Models to 1991-99 IMPROVE Aerosol Data from Brigantine Wilderness Area, NJ. **Type of Study:** Presentation the IMPROVE Steering Committee, Davis, CA. Funding Source: None mentioned **Findings and Recommendations Site and Data Specifications Source Apportionment Method** Source Apportionment Tool: Site Location(s) and Type(s): Results: Brigantine National Wildlife The average fine mass was 11 µg/m<sup>3</sup>, with the worst 20% of UNMIX the days averaging 20 µg/m<sup>3</sup> and the best 20% of the days Refuge, NJ Number of Sources/Species: averaging  $5 \mu g/m^3$ . Timeframe: 11 sources 1991-1999 Sum of the mass from the 11 sources accounts for about 90% Sources: of the measured daily fine mass. Percentages of each source Secondary sulfate (summer coal), Se Frequency: were not provided, but the largest source is secondary sulfate. source (winter coal), heavy carbon, light 24 hour averages every On the highest fine mass days, summer and winter coal are Wednesday and Saturday. carbon, secondary nitrate, oil source, Zn about 2/3 of the total mass; on low fine mass days, they are source, soil, sea salt, wood smoke, and Data Source: about 1/3 of the total mass. nitrate-Br. **IMPROVE** network Temporal Signals: Methodology: Data Description: From 1992-94 to 1997-99, total fine mass from all sources has Instead of apportioning measured fine reduced about 25%, largely due to reduction of midwest Speciated PM<sub>2.5</sub> data. mass in a single model run, four summer coal source. Winter coal increased during the same separate runs were conducted to period, possibly due to fuel switching in response to Clean Air apportion the mass of the assumed Act provisions. major mass-contributing species, chosen to be sulfates  $(1.375 \times SO_4)$ , nitrates Secondary sulfate has strong summer peak. Heavy carbon, light carbon, and soil also peak in the summer. Se sources, (1.29xNO<sub>3</sub>), carbonaceous matter (EC+1.4xOC), and crustal material secondary nitrate, Zn source, and oil source peak in the winter. (4xSi). Carbon subspecies included Total fine mass did not change from weekday to weekend, but were O1, O2, O3, O4, and EC; others oil and zinc sources were higher on weekday. Wood smoke in produced no solutions. The measured the winter and light carbon in the summer were higher on mass was not included as an input weekend. variable. No substitution was made for concentrations below MDL (only used Transboundary Transport: Trajectory analysis for summer and winter coal indicate a data above MDL). Original set of 22 common midwestern regional influence. High wood smoke sources (total from 4 runs) was reduced to 11 since redundant sources between came from distant southeast sources, consistent with summer forest fires. Oil sources seem to be located in New York City the 4 major species were highly

Study Reference: Poirot, R.L. and P.R. Wishinski (2001). Application of Combined Mathematical and Meteorological Receptor Models to 1991-99 IMPROVE Aerosol Data from Brigantine Wilderness Area, NJ. Funding Source: None mentioned Type of Study: Presentation the IMPROVE Steering Committee, Davis, CA. **Site and Data Specifications Source Apportionment Method Findings and Recommendations** correlated. These were also confirmed and Zn from northeastern NJ or eastern PA. Heavy and light carbon appear to be from the Baltimore-Washington corridor. by comparing trace elements. High soil dust days seem to link to Saharan dust events, since Other Analyses: concentrations increase down through the southern U.S. and Wind direction and ATAD back match the Al:Ca ratio for Saharan dust. trajectory analysis Recommendations: None. Upcoming Studies/Planned Work: Calculation of daily extinction, both total and from each of the sources, and comparison to the upper and lower 20% extinction days.

**Study Reference:** Investigation of Sources with Highly Time-Resolved Aerosol at the Baltimore Supersite Using PMF. Seung Park, Patrick Pancras, Yu Chang., Dawn Catino, Sarala Gazula, and John Ondov. (Poster PA 2-01.)

**Type of Study:** Poster at the American Association for Aerosol Funding Source: U.S. EPA

Research (AAAR) meeting in Charlotte, N.C., October 2002

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Clifton Park, Baltimore (urban)	Source Apportionment Tool: PMF	Results: PMF is effective for identifying possible sources from highly
Timeframe: 6-10 September 2001	Number of Sources/Species: 7 sources and 14 species	time-resolved aerosol chemical data. Ni and Se highly associated with oil and coal fired power plants. No percent contribution data for the sources was provided.
Frequency: Hourly	Sources: Coal-fired power plant, oil-fired power	Temporal Signals: None mentioned.
Data Source: Special study at the Baltimore Supersite	plant, motor vehicles/sulfate, secondary nitrate, incineration, road dust, and ferrous metal.	Transboundary Transport: None mentioned.
Data Description: Semi-continuous aerosol	Methodology: Unclear	Recommendations: None mentioned.
measurements with metal analysis (10 metals), plus nitrate, sulfate, and OC/EC.	Other Analyses: Wind direction	Upcoming Studies/Planned Work: None mentioned.

**Study Reference:** Sources of Elemental Aerosol Constituents in Pittsburgh using PMF of Highly Time-Resolved Data. Seung Park, Patrick Pancras, Sarala Gazula, and John Ondov. (Poster PA 2-05.)

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Pittsburgh (urban)  Timeframe: 16-18 July 2001  Frequency: Sub-hourly  Data Source: Special study at the Pittsburgh Supersite  Data Description: Semi-continuous aerosol measurements with metal analysis (11 metals).	Source Apportionment Tool: PMF and PCA  Number of Sources/Species: 4 sources and 11 species  Sources: PMF: urban dust, steel mill, coke or coal-fired power plant, soil dust PCA: urban dust, steel mill/coke plants, soil dust, and Pb process  Methodology: Unclear  Other Analyses: Wind direction and HYSPLIT	Results:  PMF is effective for identifying possible sources from highly time-resolved aerosol chemical data. Two peaks were observed for elements released in primary particles from high-temperature combustion sources (coking/iron processing, coal-fired power plant). No percent contribution data for the sources was provided.  Temporal Signals: None mentioned.  Transboundary Transport: None mentioned.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.

Apportionment meeting, October 2		Texas Air Research Center
Site and Data Specifications	Source Apportionment Method	Findings and Recommendations
Site Location(s) and Type(s):  3 sites in Houston, TX Aldine (urban) LaPorte (urban) HRM-3 (urban)  Timeframe: Summer 2000  Frequency: 24 hour integrated samples, every other day (organic PM) and daily (metals PM)  Data Source: Special study (TexAQS 2000)  Data Description: Organic PM from high volume virtual impactor, analyzed with GC/MS. Metals PM from sequential sampler, analyzed with XRF and ICP-MS.	Source Apportionment Tool:     CMB and PMF  Number of Sources/Species:     CMB: 7 sources     PMF: 5 sources  Sources:     CMB: diesel vehicles, gasoline     vehicles, road dust, meat cooking, wood     smoke, vegetative waxes, and fuel oil     combination.  PMF: industrial combustion, crustal     materials, road dust, wood burning, and     sea salt.  Methodology:     With CMB, used molecular markers,     including 7 alkanes, 4 petroleum     biomarkers, 7 PAHs, 2 saturated fatty     acids, 1 unsaturated fatty acid, and     levoglucosan. Also used 3 bulk     composition measures: EC, Si, Al.	Results: Only two common sources were found between PMF and CMB (road dust and wood smoke). PMF and CMB were not in good agreement. Basis of prediction (organics versus metals) is different, which could cause the poor agreement. For organics, the most important sources were diesel exhaust, gasoline vehicles, and meat cooking. For PMF, linear regression indicates crustal material was important source, followed by road dust and industrial combustion. No percent contribution data for the sources was provided.  Temporal Signals: None mentioned.  Transboundary Transport: None mentioned.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.
	Other Analyses: None.	

Study Reference: Comparing LADCO trajectory ensembles with Vt.DEC trajectory ensembles for Lye Brook (both using ATAD trajectories)

and Comparing ATAD and HYSPLIT trajectories for high source contribution days at Lye Brook. Donna Kenski.

Type of Study: Presentations at the LADCO Trajectory/Source Funding Source: None mentioned

Apportionment meeting, October 22, 2002

Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Lye Brook, VT (rural)	Source Apportionment Tool: PMF	Results: Presentation is a series of maps of the ATAD trajectory
Timeframe: March 98-March 00	Number of Sources/Species: 9 sources	ensembles (LADCO and Vt.DEC) and of the HYSPLIT trajectories for high source contribution days. ATAD and HYSPLIT trajectories give essentially the same results.
Frequency: Every 3rd day 24-hour integrated samples  Data Source: Unclear  Data Description: Aerosol Samples analyzed for	Sources: Primary coal, smelters/metal processing, limestone soil/cement, east coast incinerators, vegetative burning, secondary coal, distant soil (SW US/Sahara), east coast residual oil, and aged sea salt.	Temporal Signals: None mentioned.  Transboundary Transport: None mentioned.  Recommendations: None mentioned.
elements, SO <sub>4</sub> , NO <sub>3</sub> , EC, OC	Methodology: Unclear Other Analyses: ATAD and HYSPLIT trajectories	Upcoming Studies/Planned Work: None mentioned.

Study Reference: CMB Analysis of Urban PM2.5 Data. Donna Kenski.  Type of Study: Presentations at the LADCO Trajectory/Source Apportionment meeting, Des Plaines, IL, October 22, 2002.  Funding Source: None mentioned		
Site and Data Specifications	<b>Source Apportionment Method</b>	Findings and Recommendations
Site Location(s) and Type(s): Chicago (2 sites) (urban) Indianapolis (urban) Detroit (3 sites) (urban) Cleveland (urban) Milwaukee (urban) St. Louis (urban)  Timeframe: 1 year  Frequency: Every 3rd day integrated samples  Data Source: PM <sub>2.5</sub> speciation network  Data Description: Aerosol samples analyzed for elements, SO <sub>4</sub> , NO <sub>3</sub> , EC, OC	Source Apportionment Tool:     CMB  Number of Sources/Species:     9 sources  Sources:     Vehicles, organic, soil, lime, limestone dust, brake wear, steel production composite, secondary sulfate, nitrate.  Methodology:     Weighted Least Squares Regression  Other Analyses:     Some probability fields, unclear.	Results: Overall, source contributions are: secondary sulfate (28.5%), organic (25.7%), nitrate (16.1%), vehicles (10.6%), other (9.4), steel production composite (2.7%), and brake wear, soil, lime, and limestone dust (less than 2% each). Maps of ensemble trajectories for CMB and PMF are also included but no results presented.  Temporal Signals: None mentioned.  Transboundary Transport: None mentioned.  Recommendations: None mentioned.  Upcoming Studies/Planned Work: None mentioned.

Table A-4. Source matrix from published apportionment studies by approximate geography

				Sulfate/ Coal		Mobile			_	nass ning	Industrial				Cri	ustal a Salt	ind	Miscellaneous		
Location or Nearest City	Reference	Total Mass (µg/m³)	Regional transport secondary	Local	General	Secondary OC/general	Diesel	Gasoline	Wood smoke	Vegetative burning	Smelter	Incinerator	Oil/diesel combustion	Other/ general	Crustal/dirt/ soil	Road salt	Sea salt	Not Identified	Other	Other defined
NORTHEAST & MIDATLANTIC																				
Acadia Natl Pk, ME	Coutant et al. 2002	7.5	2.3			3.9			0.3			0.4	0.0		0.1		0.4	0.1		
Lye Brook, VT	Coutant et al. 2002	7.6	3.6			0.4				2.4	0.5	0.2			0.5	0.1				
Underhill, VT	Poirot et al. 2001 (PMF)	7.9	4.8						1.2		0.1		0.6	0.9	0.3	0.0				
Underhill, VT	Poirot et al. 2001 (UNMIX)	8.4	5.8						1.3		0.1		0.7	0.3	0.2					
Underhill, VT	Polissar et al. 2001	6.4	3.7						1.0		0.2		0.4	0.6	0.3	0.1		0.3		
Underhill, VT	Song et al. 2001	8.0	4.9						1.2		0.2	0.6	0.5	0.2	0.3	0.0	0.1			
Bronx, NY	Coutant et al. 2003	16.1	5.3		4.1	2.5							1.2	1.8	1.0		0.3			
Connecticut Hill, NY	Coutant et al. 2002	19.0	5.5			1.5							0.1		0.1			11.8		
Brigantine, NJ	Coutant et al. 2002	11.6	5.7			3.5							1.3	0.3	0.1		0.7	0.0		
Brigantine, NJ	Lee, JH et al. 2003a	11.4	7.9			1.8						0.2	0.3		0.2		0.9			
Brigantine, NJ	Song et al. 2001	11.4	7.1		0.9	0.6			0.9			0.5	0.2		0.1		0.9			
Arendtsville, PA	Coutant et al. 2002	22.5	6.2			2.0				0.1			0.0		0.1	0.0		14.0	0.1	Smudge pots
M.K.Goddard, PA	Coutant et al. 2002	19.5	5.8			4.2				0.4		0.4	0.3		8.3			0.0		0 1
Baltimore-Wash. corridor	Chen et al. 2002	13.	5.3	0.9	2.0	1.8			1.6		0.2									
Washington, DC	Coutant et al. 2002	17.9	7.5			6.6				0.3			1.8		1.4		0.3			
Washington, DC	Coutant et al. 2003	16.7	7.7		1.2	4.7			1.1						1.5				0.5	Fireworks
Washington, DC	Song et al. 2001	17.6	10.0		3.5	1.6						0.9	0.6		0.5		0.6			
Washington, DC	Kim and Hopke 2003	17.9	10.7		1.6		0.4	3.8				0.7	0.4		0.4		0.4			
Jeff./James River, VA	Coutant et al. 2002	14.7	7.2			3.7			2.2			0.3	0.4	0.5	0.5					
Shenandoah, VA	Coutant et al. 2002	11.8	4.5			3.2				3.5					0.2		0.3	0.1		
Dolly Sods, WV	Coutant et al. 2002	12.7	5.0			4.5				0.7		0.2	0.2		0.8		1.2			
MIDWEST																				
Toronto, ON	Lee, PKH et al. 2003b	12.7	3.3		4.6	1.9					0.1		0.1	0.0	1.3	1.0			0.0	Construction
Quaker City, OH	Coutant et al. 2002	20.5	6.3			2.0				0.0					8.8			3.4		
Livonia, IN	Coutant et al. 2002	20.1	6.3			3.9				0.1	0.2	1.9			7.8					
Mammoth Cave, KY	Coutant et al. 2002	16.0	4.9			6.1			3.0				0.7	0.1	0.8	0.3				
Great Smoky, TN	Coutant et al. 2002	13.4	4.8			6.6				0.7		0.3	0.2		0.6		0.2	0.1		
Indianapolis, IN	Coutant et al. 2003	17.3	8.7		3.6	3.2			0.3						0.5		0.5		0.7	Fireworks
Bondville, IL	Coutant et al. 2002	18.7	5.4			0.3				0.8		0.1			0.1			12.0		
St. Louis, MO	Coutant et al. 2003	17.2	5.7		5.0	2.9					2.2				1.4					
Milwaukee, WI	Coutant et al. 2003	14.5	4.5		4.1	2.5								2.7	0.3				0.4	Fireworks
Boundary Waters, MN	Coutant et al. 2002	5.4	2.4			2.2				0.2		0.2		0.2	0.2	0.1				
SOUTHEAST & SOUTH																				
Charlotte, NC	Coutant et al. 2003	16.2	5.7		1.2	3.9					0.7		1.9		0.6		0.1		0.5	Fireworks

			Sulfate/ Coal		Nitr ate	Monie			_	nass ning		Industrial				Crustal and Salt			Miscellaneous		
Location or Nearest City	Reference	Total Mass (µg/m³)	Regional transport secondary	Local	General	Secondary OC/general	Diesel	Gasoline	Wood	Vegetative burning	Smelter	Incinerator	Oil/diesel combustion	Other/ general	Crustal/dirt/ soil	Road salt	Sea salt	Not Identified	Other	Other defined	
Atlanta, GA	Kim et al. 2003a	18.3	10.2		1.6		2.0	1.1	1.3		0.5			0.7	0.5				0.5	Railroad	
Atlanta, GA	Kim et al. 2003b	18.0	10.1		1.6		2.0	2.7	0.5		0.5			0.2	0.4						
Atlanta, GA	Kim et al. 2002a	18.0	10.1		1.3	4.0			2.0		0.1			0.4	0.2						
Birmingham, AL	Coutant et al. 2003	19.5	7.3		1.8	6.5								1.5	1.3				1.2	Fireworks	
Houston, TX	Coutant et al. 2003	14.2	5.5			6.2								0.9	8.0		0.3		0.5	Fireworks	
WEST																					
Phoenix, AZ	Lewis et al. 2002	12.6	2.4				2.0	4.2		1.3					2.8						
Spokane, WA	Kim et al. 2002c	12.1	2.3		1.1	1.3				5.3	0.4	0.7			1.1						
Seattle, WA	Maykut et al. 2003 (CMB)	8.9	1.5		0.4	3.9				1.4				0.6	0.4		0.6				
Seattle, WA	Maykut et al. 2003 (UNMIX)	8.9					1.7	0.8		3.3			1.3		0.5		1.1				
Seattle, WA	Maykut et al. 2003 (PMF)	8.9	1.6				1.6	0.4		2.5			0.9		1.2		0.3		0.4	Na-rich	

Notes: All values are in µg/m3. Sources greater than 20 percent but less than 40 percent of the total mass are shaded yellow; sources greater than 40 percent are shaded pink.

In order to present a concise overview, some sources were renamed and/or combined in some studies. Some values were calculated based on reported percentages. Sum of sources may not equal the total due to rounding and/or modeling limitations. Refer to descriptions in Table A-3 for exact source names, number of sources, and values.

Studies not included:

Battye (2002) presented back trajectory analysis only with no sources identified.

Wishinski and Poirot (1986) measured visibility and did not report concentrations or mass.

Long (2002) focused on 3 one-week "event" studies (e.g., forest fires, Olympic Games) and did not present overall results.

Zheng et al. (2002) presented concentration data for four one-month sampling periods for 8 sites but no annual average by site and source.

Lowenthal & Rahn (1989) and Rahn & Lowenthal (1984) are early studies that identified sources as regional areas.

Kim et al. (2002b), Liu et al. (2003b), Liu et al. (2003b), Ramadan et al. (2000), Ramadan et al. (2003) did not provide sufficient concentration and/or percent

mass values to complete this table.

Chow and Watson (2002) was a summary of other studies.